Criteria Pollutants — National Trends

This chapter presents national and regional trends for each of the six criteria pollutants for which the U.S. Environmental Protection Agency (EPA) has established National Ambient Air Quality Standards (NAAQS): carbon monoxide (CO), lead (Pb), nitrogen dioxide (NO_2), ozone (O_3), particulate matter (PM), and sulfur dioxide (SO_2). Table 2-1 lists the NAAQS for each pollutant in terms of the level and averaging time of the standard used to evaluate compliance.

There are two types of standards: primary and secondary. Primary standards protect against adverse human health effects, whereas secondary standards protect against welfare effects such as damage to crops, ecosystems, vegetation, and buildings, as well as decreased visibility. There are primary standards for all of the criteria pollutants. Some pollutants (PM and SO_2) have primary standards for both long-term (annual average) and short-term (24 hours or less) averaging times. Short-term standards most directly protect people from adverse health effects associated with peak short-term exposures to air pollution, whereas long-term standards can protect people from adverse health effects associated with short- and long-term exposures to air pollution.

Table 2-1. NAAQS in Effect as of December 2002

Pollutant	Primary Standard (Health-Related)		Secondary Standard (Welfare-Related)	
	Type of Average	Standard Level Concentration[a]	Type of Average	Standard Level Concentration[a]
CO	8-hour[b]	9 ppm (10 mg/m^3)	No Secondary Standard	
	1-hour[b]	35 ppm (40 mg/m^3)	No Secondary Standard	
Pb	Maximum Quarterly Average	1.5 µg/m^3	Same as Primary Standard	
NO$_2$	Annual Arithmetic Mean	0.053 ppm (100 µg/m^3)	Same as Primary Standard	
O$_3$	Maximum Daily 1-hour Average[c]	0.12 ppm (235 µg/m^3)	Same as Primary Standard	
	4th Maximum Daily[d] 8-hour Average	0.08 ppm (157 µg/m^3)	Same as Primary Standard	
PM$_{10}$	Annual Arithmetic Mean	50 µg/m^3	Same as Primary Standard	
	24-hour[e]	150 µg/m^3	Same as Primary Standard	
PM$_{2.5}$	Annual Arithmetic Mean[f]	15 µg/m^3	Same as Primary Standard	
	24-hour[g]	65 µg/m^3	Same as Primary Standard	
SO$_2$	Annual Arithmetic Mean	0.03 ppm (80 µg/m^3)	3-hour[b]	0.50 ppm (1,300 µg/m^3)
	24-hour[b]	0.14 ppm (365 µg/m^3)		

[a] Parenthetical value is an approximately equivalent concentration. (See *40 CFR* Part 50.)
[b] Not to be exceeded more than once per year.
[c] The standard is attained when the expected number of days per calendar year with maximum hourly average concentrations above 0.12 ppm is equal to or less than 1, as determined according to Appendix H of the Ozone NAAQS.
[d] Three-year average of the annual 4th highest daily maximum 8-hour average concentration.
[e] The short-term (24-hour) standard of 150 µg/m^3 is not to be exceeded more than once per year on average over 3 years.
[f] Spatially averaged over designated monitors.
[g] The form is the 98th percentile.

Secondary standards have been established for each criteria pollutant except CO. Secondary standards are identical to the primary standards, with the exception of the one for SO_2. As Figure 2-1 shows, approximately 146 million people in the United States reside in counties that did not meet the primary standard for at least one of the criteria pollutants for the single year 2002.

Figure 2-1. Number of people living in counties with air quality concentrations above the level of NAAQS in 2002.
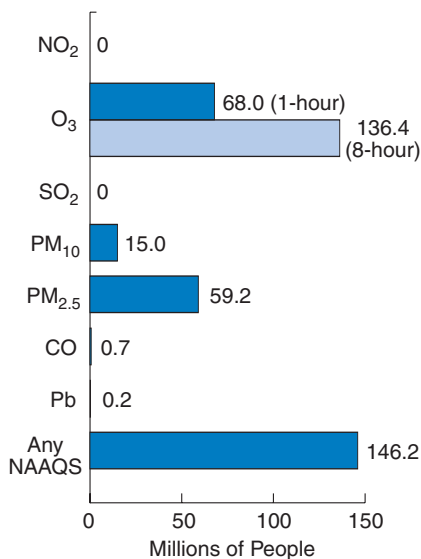
The trends information presented in this chapter is based on two types of data: ambient concentrations and emissions estimates. Ambient concentrations are measurements of pollutant concentrations in the ambient air from monitoring sites across the country. This year's report contains trends data accumulated between 1983 and 2002 on the criteria pollutants at thousands of monitoring stations located throughout the United States. For some pollutants, 2002 data are provided; for other pollutants (e.g., lead), 2001 data are

reported. In each case, the most recent, complete data are used, with the relevant years clearly noted. The trends presented here are derived from the composite average of these direct measurements. The averaging times and air quality statistics used in the trends calculations relate directly to the NAAQS.

The second type of data presented in this chapter are national emissions estimates. These are based largely on engineering calculations of the amounts and kinds of pollutants emitted by automobiles, factories, and other sources over a given period. In addition, some emissions estimates are based on measurements from continuous emissions monitors (CEMs) that have been installed at major electric utilities to measure actual emissions. The emissions data summarized in this chapter and in Appendix A were obtained from the National Emission Inventory data located at **http://www.epa.gov/ttn/chief**.

Methods for estimating emissions continue to evolve. For example, the emissions data presented here reflect the use of new models for estimating volatile organic compounds (VOCs), nitrogen oxides (NO_x), and CO emissions from highway vehicles and nonroad engines. Also, emissions from wildfires and prescribed burning have not been projected for 2002 for PM, CO, and VOCs, due to the high degree of uncertainty in predicting emissions for these fires. For a complete description of the methodology changes for calculating emissions, see Appendix B.

Changes in ambient concentrations do not always match changes in national emissions estimates, for several reasons. First, because most monitors are positioned in urban, population-oriented locales, air

quality trends are more likely to track changes in urban emissions rather than changes in total national emissions. Urban emissions are generally dominated by mobile sources, whereas total emissions in rural areas may be dominated by large stationary sources such as power plants and smelters.

Second, emissions for some pollutants are calculated or measured in a different form than the primary air pollutant. For example, concentrations of O_3 are caused by VOC emissions as well as NO_x emissions.

Third, the amount of some pollutants measured at monitoring locations depends on what chemical reactions, if any, occur in the atmosphere during the time it takes the pollutant to travel from its source to the monitoring station.

Fourth, meteorological conditions often control the formation and buildup of pollutants in the ambient air. For example, peak ozone concentrations typically occur during hot, dry, stagnant summertime conditions. CO is predominantly a cold weather problem. Also, the amount of rainfall can affect particulate matter levels.

Fifth, emissions estimates have uncertainties and may not reflect actual emissions. In some cases, estimation methods are not consistent across all years presented in this report.

For a more detailed discussion of the methodology used to compute the trend statistics in this chapter, please refer to Appendix B.

Carbon Monoxide

Air Quality Concentrations		
1983–02	65%	decrease
1993–02	42%	decrease

Emissions		
1983–02	41%	decrease
1993–02	21%	decrease

Worth Noting

- Nationally, carbon monoxide (CO) levels for 2002 are the lowest recorded in the past 20 years and improvement is consistent across all regions of the country.

- All of the original 42 areas designated nonattainment for the 8-hour CO NAAQS in 1991 met the CO NAAQS in 2001–2002.

- However, three additional areas failed to meet the CO NAAQS in 2001–2002.

Nature and Sources

Carbon monoxide is a colorless and odorless gas, formed when carbon in fuel is not burned completely. It is a component of motor vehicle exhaust, which contributes about 60 percent of all CO emissions nationwide. Nonroad vehicles account for the remaining CO emissions from transportation sources. High concentrations of CO generally occur in areas with heavy traffic congestion. In cities, as much as 95 percent of all CO emissions may come from automobile exhaust. Other sources of CO emissions include industrial processes, nontransportation fuel combustion, and natural sources such as wildfires. Peak CO concentrations typically occur during the colder months of the year when CO automotive emissions are greater and nighttime inversion conditions (where air pollutants are trapped near the ground beneath a layer of warm air) are more frequent.

Health Effects

CO enters the bloodstream through the lungs and reduces oxygen delivery to the body's organs and tissues. The health threat from levels of CO sometimes found in the ambient air is most serious for those who suffer from cardiovascular disease such as angina pectoris. At much higher levels of exposure not commonly found in ambient air, CO can be poisonous, and even healthy individuals may be affected. Visual impairment, reduced work capacity, reduced manual dexterity, poor learning ability, and difficulty in performing complex tasks are all associated with exposure to elevated CO levels.

Primary Standards

There are two primary NAAQS for ambient CO: a 1-hour average of 35 ppm and an 8-hour average of 9 ppm. These concentrations are not to be exceeded more than once per year. There currently are no secondary standards for CO.

National Air Quality Trends

Nationally, CO concentrations have consistently declined over the past 20 years. Figure 2-2 reveals a 65 percent improvement in composite average ambient CO concentrations from 1983 to 2002 and a 42 percent reduction over the past 10 years.[1] Following an upturn in 1994, the nation experienced year-to-year reductions in peak 8-hour CO concentrations through the remainder of the decade. In fact, the 2002 CO levels were the lowest recorded during the past 20 years. Exceedances of the 8-hour CO NAAQS (which are simply a count of the number of times the level of the standard is exceeded) have declined. In fact, all of the original 42 areas designated nonattainment for the 8-hour CO NAAQS in 1991 met the CO NAAQS in 2001–2002. However, three additional areas failed to meet the CO NAAQS in 2001–2002. This improvement occurred despite a 23 percent increase in vehicle miles traveled in the United States during the past 10 years.

Figure 2-2. CO air quality, 1983–2002, based on annual second maximum 8-hour average.

1983–02: 65% decrease
1993–02: 42% decrease
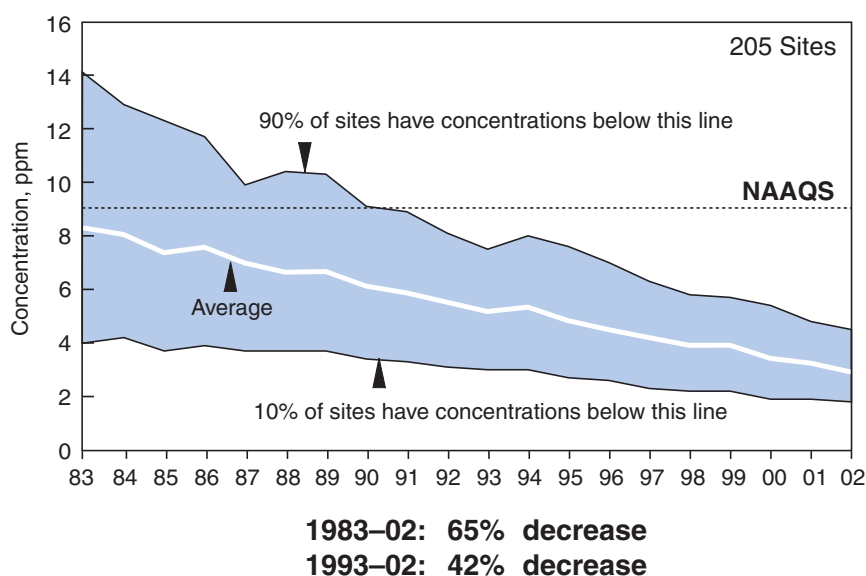
Long-term reductions in ambient CO concentrations have been measured across all monitoring environments—rural, suburban, and urban sites. Figure 2-3 shows that, on average, urban monitoring sites record higher CO concentrations than do suburban sites, with the lowest levels found at four rural sites. During the past 20 years, the 8-hour CO concentrations decreased 44 percent at 4 rural monitoring sites, 60 percent at 89 suburban sites, and 63 percent at 116 urban sites.

Regional Air Quality Trends

The map in Figure 2-4 shows regional trends in ambient CO concentrations during the past 20 years, 1982 to 2001. All 10 EPA Regions recorded 20-year improvements in CO levels as measured by the regional composite mean concentrations. Significant 20-year concentration reductions of 50 percent or more were evidenced across the nation.

National Emissions Trends

Figure 2-5 shows that the transportation category, composed of onroad and nonroad sources, accounted for 82 percent of the nation's total CO

Figure 2-3. Trend in second maximum nonoverlapping 8-hour average CO concentrations by type of location, 1982–2001.
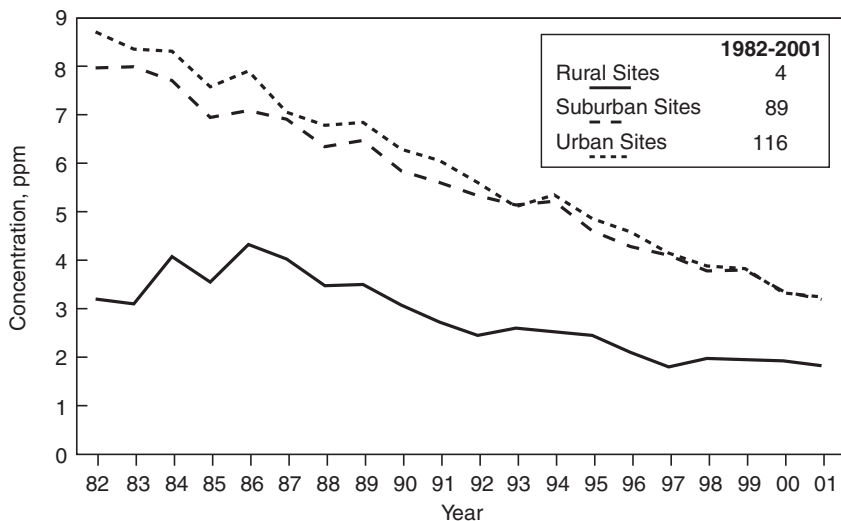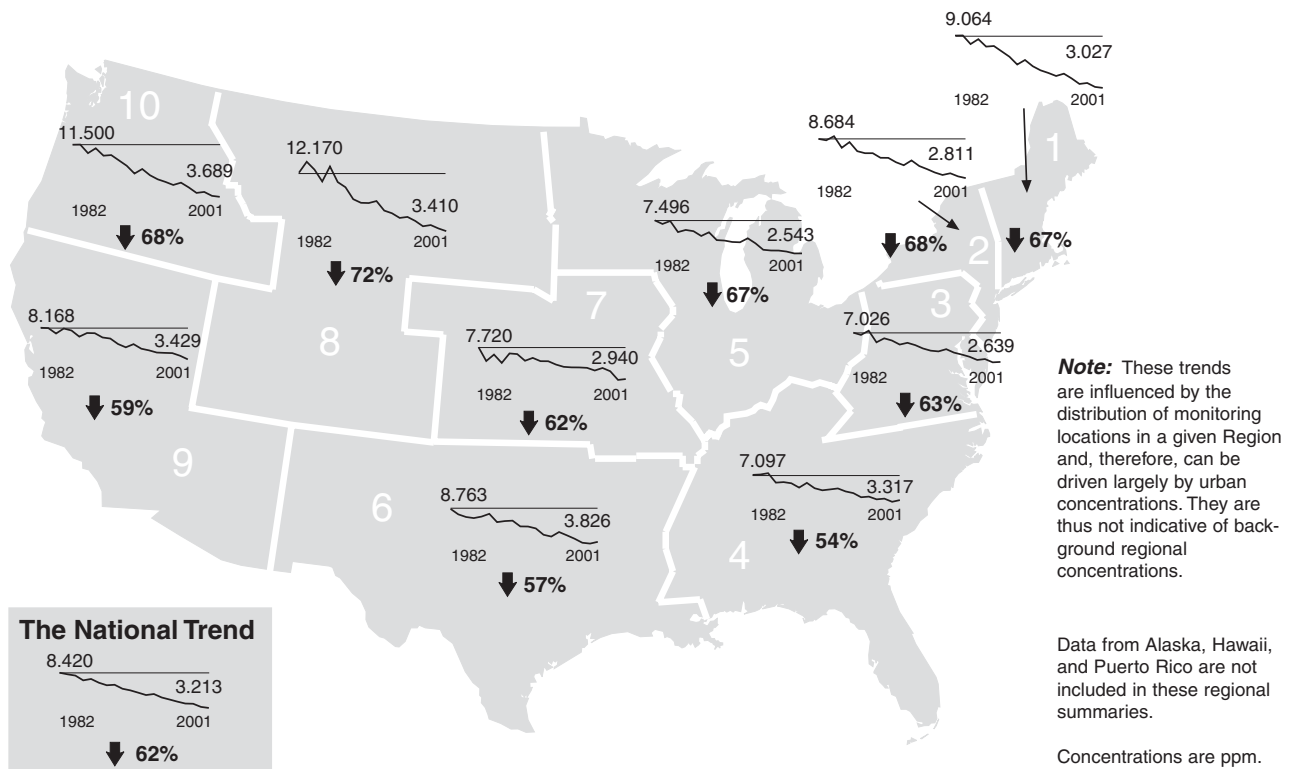

	1982-2001
Rural Sites	4
Suburban Sites	89
Urban Sites	116

Figure 2-4. Trend in CO second maximum nonoverlapping 8-hour concentrations by EPA Region, 1982–2001.

Note: These trends are influenced by the distribution of monitoring locations in a given Region and, therefore, can be driven largely by urban concentrations. They are thus not indicative of background regional concentrations.

Data from Alaska, Hawaii, and Puerto Rico are not included in these regional summaries.

Concentrations are ppm.

emissions in 2002. Figure 2-6 presents the broad geographic distributions of 2001 CO emissions based on the tonnage per square mile for each county. This visualization clearly shows that the eastern third of the country and the West Coast emitted more CO (on a density basis) than

did the western two-thirds of the continental United States. As a result of automotive emissions control programs, CO emission have decreased 41 percent the past 20 years (1983 to 2002) and 21 percent in the past 10 years (1993 to 2002) despite a 155 percent increase in VMT since 1970

(see Figure 2-7). However, emissions from all transportation sources have decreased only 10 percent over the same period, primarily due to an increase in offroad emissions that has offset the gains realized in reductions of onroad vehicle emissions.

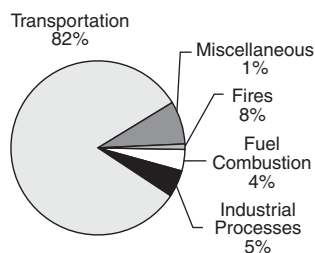**Figure 2-5**. CO emissions by source category, 2002.

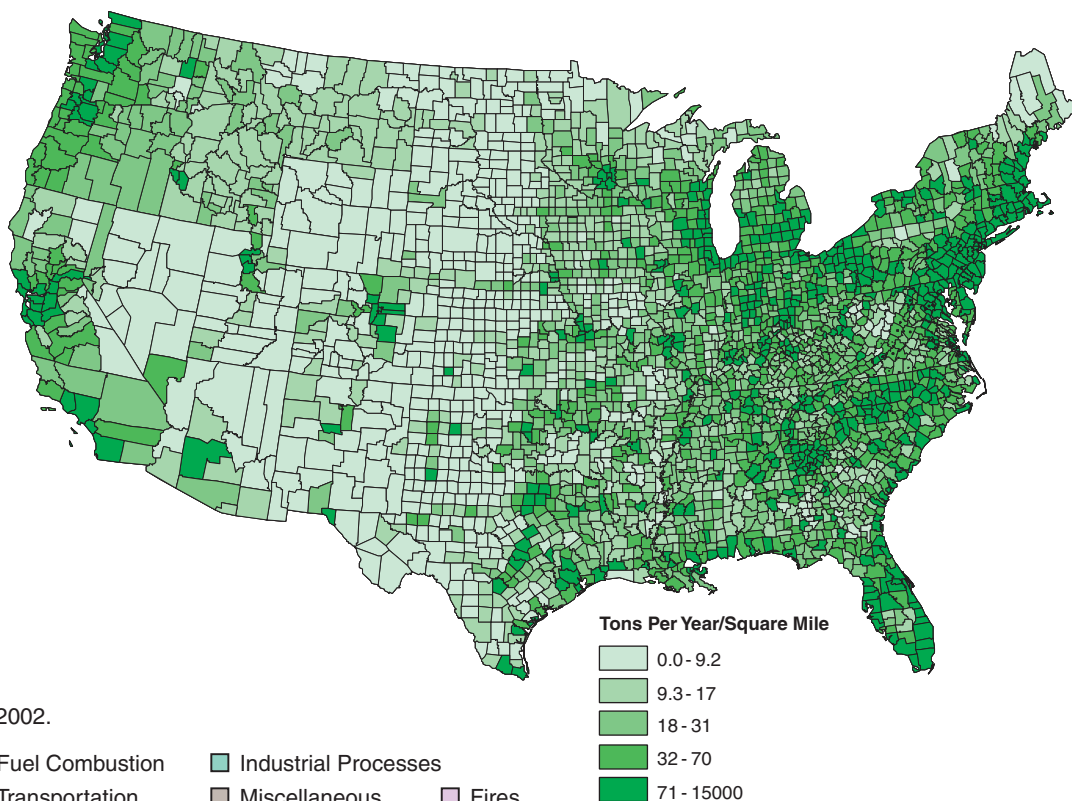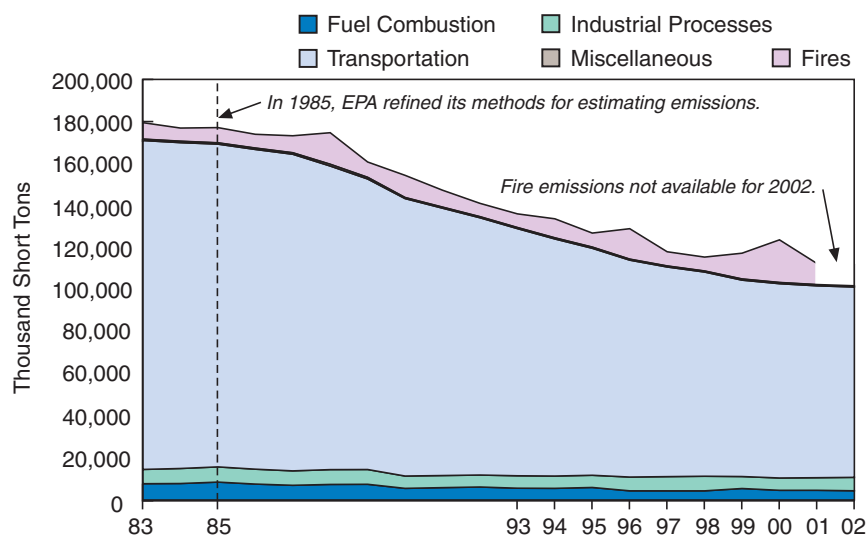**Figure 2-6**. Density map of 2001 CO emissions, by county.

Tons Per Year/Square Mile

	0.0 - 9.2
	9.3 - 17
	18 - 31
	32 - 70
	71 - 15000

Figure 2-7. CO emissions, 1983–2002.

1983–02: **41% decrease**[a]
1993–02: **21% decrease**

Note: Emission estimation methods and data sources have evolved over time, resulting in some inconsistency in estimates in different years. In the methods used for this report, the significant changes have occurred between 1984 and 1986 and between 1995 and 1996, although not all source types were affected. More explanation is provided in Appendix B.

[a] Emissions trends data are not available for 1983; thus, the 20-year trend was interpolated based on emissions data for 1980 and 1985.

Table 2-2 lists some of the major milestones in the control of emissions from automobiles, starting with the Clean Air Act (the Act) of 1970. At the national level, these measures, which have led to reductions in emissions of CO as well as other pollutants, include establishing national standards for tailpipe emissions, new vehicle technologies, and clean fuels programs. State and local emissions reduction measures include inspection and maintenance (I/M) programs and transportation management programs.

In the area of clean fuels, the 1990 Clean Air Act Amendments (1990 Amendments) require oxygenated gasoline programs in several regions of the country during the winter months. Under the program regulations, a minimum oxygen content (2.7 percent by weight) is required in gasoline to ensure more complete fuel combustion.[2,3] Of the 36 CO nonattainment areas that initially implemented the program in 1992, 15 areas participated in the program during 2000.[4]

2001 Air Quality Status

The map in Figure 2-8 shows the variations in CO concentrations across the country in 2001. The air quality indicator is the largest annual second maximum 8-hour CO concentration measured at any site in each county. The bar chart to the left of the map displays the number of people living in counties within each concentration range. The colors on the map and bar chart in Figure 2-8 correspond to the colors of the concentration ranges displayed in the map legend. The only areas not meeting the 8-hour CO NAAQS in 2001–2002 are Birmingham, AL, Calexico, CA, and Weirton, WV.

Table 2-2. Milestones in motor vehicle emission control.

1970 New Clean Air Act sets auto emissions standards.	1989 Fuel volatility limits are set for Reid Vapor Pressure (RVP).
1971 Charcoal canisters appear to meet evaporative standards.	1990 The 1990 Amendments set new tailpipe standards.
1973 Emission gas recycle (EGR) valves appear to meet NO_x standards.	1992 Oxyfuel introduced in cities with high CO levels.
1974 Fuel economy standards are set.	1993 Limits set on sulfur content of diesel fuel.
1975 The first catalytic converters appear for hydrocarbon, CO. Unleaded gas appears for use in catalyst-equipped cars.	1994 Phase-in begins of new vehicle standards and technologies.
1981 Three-way catalysts with onboard computers and O_2 sensors appear.	1995 Onboard diagnostic systems in 1996 model-year cars.
1983 Inspection and maintenance programs (I/M) programs are established in 64 cities.	1995 Phase I Federal Reformulated Gasoline sales begin in worst ozone nonattainment areas.
	1998 Sales of 1999 model-year California emissions-equipped vehicles begin in the Northeast.

Figure 2-8. Highest second maximum nonoverlapping 8-hour average CO concentration by county, 2001.
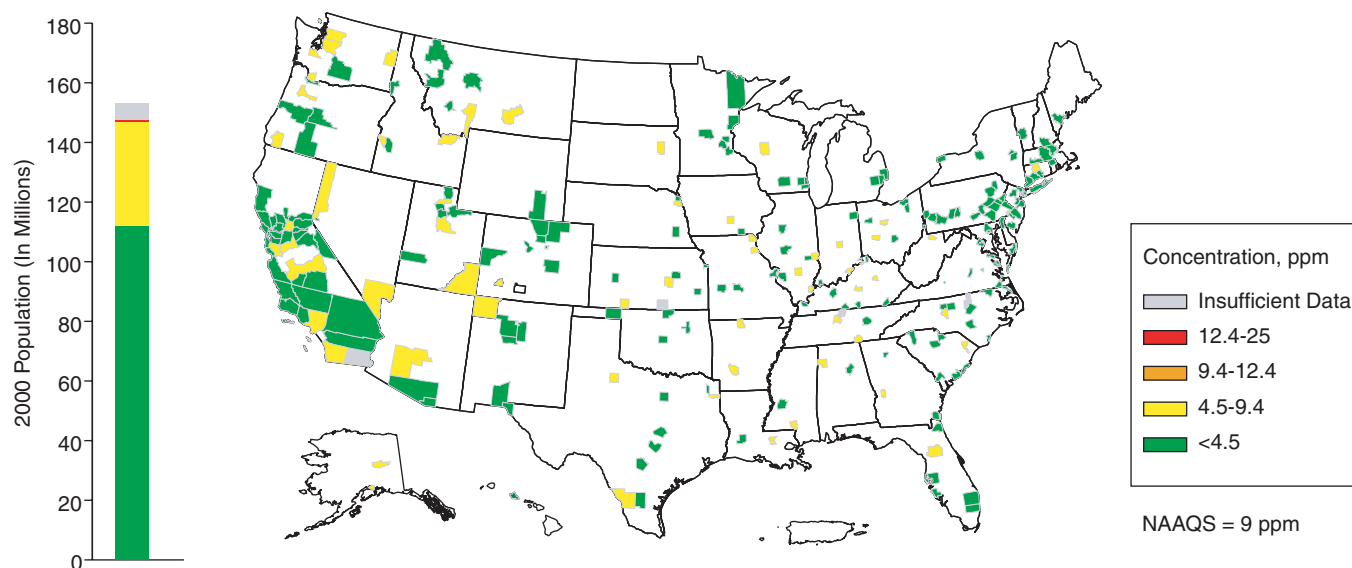
Lead

Air Quality Concentrations		
1983–02	94%	decrease
1993–02	57%	decrease
Emissions		
1982–02	93%	decrease
1993–02	5%	decrease

Worth Noting

- The lead (Pb) monitoring strategy now focuses on emissions from point sources since large reductions in long-term Pb emissions from transportation sources have occurred due to phase-out of leaded gasoline.

Nature and Sources

In the past, automotive sources were the major contributor of lead emissions to the atmosphere. As a result of EPA's regulatory efforts to reduce the content of lead in gasoline, however, the contribution of air emissions of lead from the transportation sector, and particularly the automotive sector, has greatly declined over the past two decades. Today, industrial processes, primarily metals processing, are the major source of lead emissions to the atmosphere. The highest air concentrations of lead are usually found in the vicinity of smelters and battery manufacturers.

Health and Environmental Effects

Exposure to lead occurs through ingestion of lead in food, water, soil, or dust and through inhalation. It accumulates in the blood, bones, and soft tissues. Lead can also adversely affect the kidneys, liver, nervous system, and other organs. Excessive exposure to lead may cause neurological impairments such as seizures,

mental retardation, and/or behavioral disorders. Even at low doses, Pb exposure is associated with changes in fundamental enzymatic, energy transfer, and homeostatic mechanisms in the human body. Additionally, even low levels of Pb exposure may cause central nervous system damage in fetuses and children. Recent studies show that neurobehavioral changes may result from Pb exposure during the child's first years of life and that lead may be a factor in high blood pressure and subsequent heart disease.

Airborne lead can also have adverse impacts on the environment. Wild and domestic grazing animals may ingest lead that has deposited on plant or soil surfaces or that has been absorbed by plants through leaves or roots. Animals, however, do not appear to be more susceptible or more sensitive to adverse effects from lead than are humans. Therefore, the secondary standard for lead is identical to the primary standard.

At relatively low concentrations $(2–10 \, \mu g/m^3)$, lead can inhibit plant growth and result in a shift to more tolerant plant species growing near roadsides and stationary source emissions. Although the majority of soil lead becomes bound so that it is insoluble, immobile, and biologically unavailable, elevated soil Pb concentrations have been observed to cause shifts in the microbial community (fungi and bacteria), reduced numbers of invertebrates, and reduced decomposition and nitrification rates and has altered other soil parameters. Because lead remains in the soil, soil concentrations continue to build over time, even when deposition rates are low. Thus, another concern is that acid precipitation may be increasing the mobility and bioavailability of soil lead in some places.

Lead enters water systems mainly through urban runoff, sewage effluents, and industrial waste streams. Most of this lead is rapidly complexed and bound in the sediment. However, water Pb concentrations can reach levels that are associated with increased mortality and impaired reproduction in aquatic invertebrates and blood and neurological changes in fish. Because of these effects, there continue to be implications for the long-term impact of lead on ecosystem function and stability. (See also Chapter 5 in this report as well as the December 1990 Office of Air Quality Planning and Standards Staff Paper [EPA-450/2-89-022].)

Primary and Secondary Standards

The primary as well as secondary NAAQS for lead is a quarterly average concentration not to exceed $1.5 \, \mu g/m^3$.

National Air Quality Trends

The statistic used to track ambient lead air quality is the maximum quarterly mean concentration for each year. From 1982 to 2001, a total of 39 ambient Pb monitors met the trends data completeness criteria, and a total of 96 ambient Pb monitors met the trends data completeness criteria for the 10-year period from 1992 to 2001. Point-source-oriented monitoring data were omitted from all ambient trends analysis presented in this section to avoid masking the underlying urban trends.

Figure 2-9 indicates that between 1993 and 2002, maximum quarterly average Pb concentrations decreased 57 percent at population-oriented monitors. Between 1999 and 2002, national average Pb concentrations (approaching the minimum detectable level) remained unchanged.

The effect of the conversion to unleaded gasoline usage in vehicles on ambient Pb concentrations is most evident when viewed over a longer period, such as that illustrated in Figure 2-9. Between 1983 and 2002, ambient monitor data indicate that concentrations of lead declined 94 percent. This large decline tracks well with overall Pb emissions, which also declined approximately 93 percent between 1983 and 2002.

Figure 2-10 examines urban, rural, and suburban 20-year trends separately. The overall downward trend in Pb concentrations can be noted for all locations from 1982 to 2001.

National Emission Trends

For stationary sources, Pb emissions for past trends reports have been estimated for fuel combustion and industrial sources based on current data for national activity, but with emission factor and control efficiency estimates that have not been updated with any new information in many years. When gasoline contained lead, mobile sources were by far the largest contributor to Pb emissions, and approximations for stationary sources did not introduce much uncertainty into the understanding of the total emissions trend. Now, most lead is emitted by industrial facilities, particularly by primary and secondary metals processing plants. Moreover, many of these facilities have been the focus of control and compliance efforts in recent years. There are also some issues of possible double counting and inventory gaps.

For example, about 10 percent of Pb emissions estimated in previous reports were from miscellaneous fuel combustion, the only element of which is the combustion of used motor oil containing lead picked up from gasoline. This estimate should

be viewed with caution, as the reduction factor of 90 percent used for this source category to reflect the end of leaded gasoline for highway use seems inconsistent with a much greater reduction factor used for exhaust emissions from vehicles. Also, the emission estimates for the sources that burn this fuel (e.g., cement kilns) may double count some of the Pb emissions. Conversely, the estimate of zero Pb emissions from

nonroad gasoline engines is inconsistent with the assumption for highway vehicles that cross-contamination with leaded aviation gasoline causes unleaded fuel to still have small amounts of Pb content on average. Aviation gasoline is not regulated for Pb content and can use significant amounts of lead to comply with octane requirements.

EPA believes that the uncertainties in the past top-down approach for

Figure 2-9. Pb air quality, 1983–2002, based on annual maximum quarterly average.

1983–02: 94% decrease
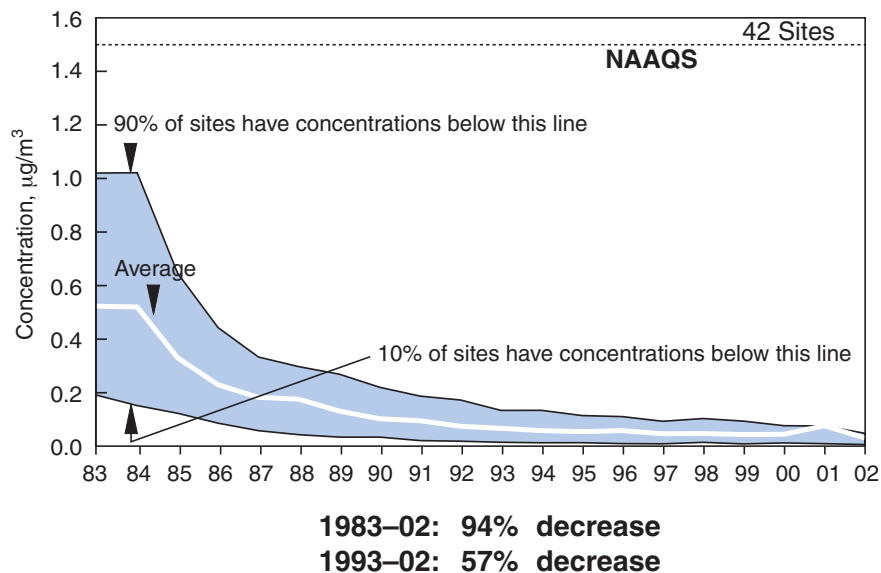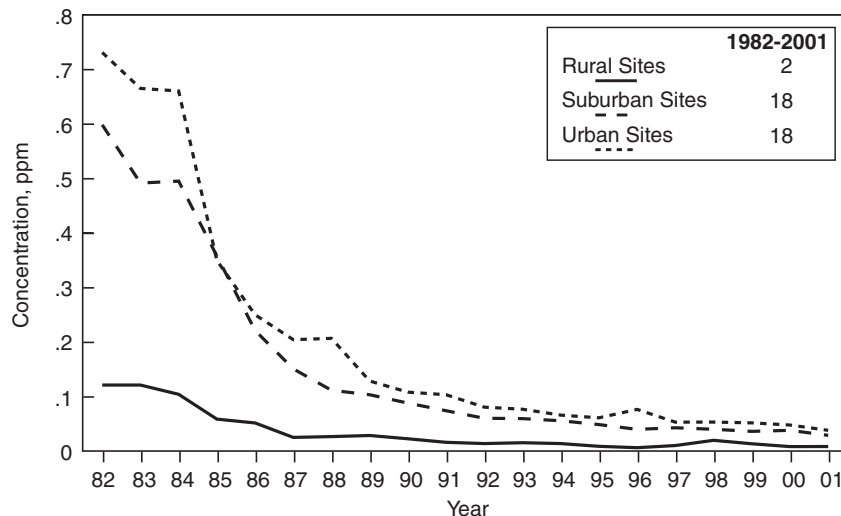**1993–02: 57% decrease**

Figure 2-10. Maximum quarterly mean Pb concentration trends by location (excluding sites designated as point-source oriented), 1982–2001.

fuel combustion and industrial sources are greater than the actual year-to-year variation in emissions. Consequently, we have not repeated it for this report. The Pb emission estimates for these sources presented here are the same as in the *1999 National Air Quality and Emissions Trends Report*, with the previous estimates for 2000 repeated for 2001. Lead emissions for transportation sources have been adjusted for activity changes.

The preferred approach for estimating Pb emissions is to make facility-specific estimates for the source types with significant emissions, reflecting the best information on fuel and ore Pb content, control equipment, and throughput. Ideally, emission tests would be conducted. For the single year of 1996, EPA collected as many such estimates as possible from state/local air agencies, the Toxics Release Inventory, and from EPA studies in preparation for the promulgation of emission standards. A comparison of these estimates to the earlier top-down estimates suggests that Pb emissions from coal-fired utilities may have been higher in 1996 than stated in this report, depending on whether a few states have correctly estimated such emissions. Emissions of lead from other industrial sources in 1996 were somewhat lower than reported in this document for that year.

Regardless of these uncertainties, the long-term trend in Pb emissions is very clear. Because of the phase-out of leaded gasoline, Pb emissions (and concentrations) decreased sharply during the 1980s and early 1990s. There was an approximate decrease in Pb emissions of 93 percent from 1982 to 1991. Figure 2-11 indicates that total Pb emissions have stayed about the same from 1991 on. The large ambient and emission reductions in lead going

from 1982 to 1991 can be largely attributed to the phasing out of leaded gasoline for automobiles. Relative to levels in the 1970s, Pb emissions in the past 10 years have been essentially constant.

Figure 2-12 shows that industrial processes were the major source of Pb

emissions in 2001, accounting for 78 percent of the total. The transportation sector (which includes both onroad and nonroad sources) now accounts for only 12 percent of the total 2001 Pb emissions, with most of that coming from aircraft.

Figure 2-11. Pb emissions, 1982–2002.
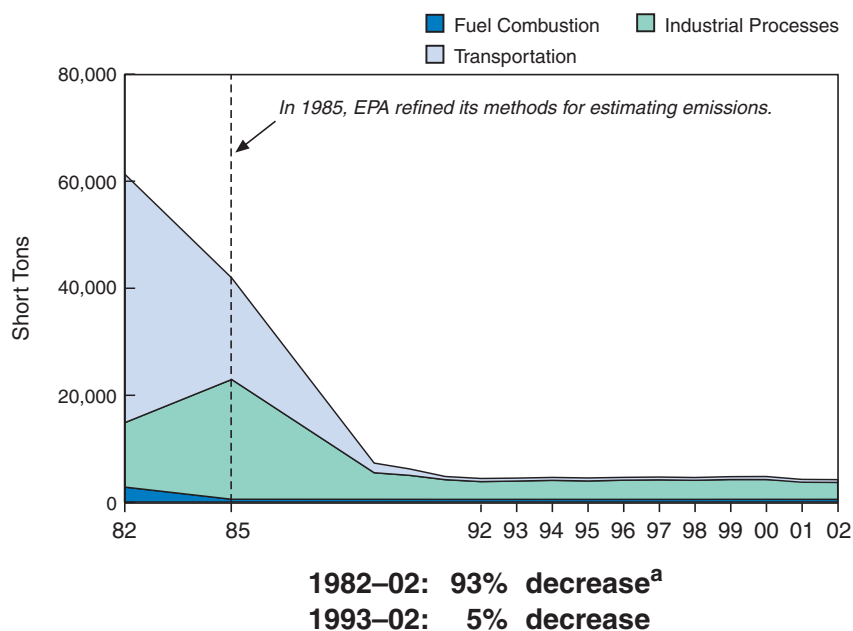
1982–02: 93% decrease[a]
1993–02:   5% decrease

Note: Emission estimation methods and data sources have evolved over time, resulting in some inconsistency in estimates in different years. In the methods used for this report, the significant changes have occurred between 1984 and 1986, and between 1995 and 1996, although not all source types were affected. More explanation is provided in Appendix B.

[a] Emissions trends data are not available for 1982; thus, the 20-year trend was interpolated based on emissions data for 1980 and 1985.

Figure 2-12. Pb emissions by source category, 2001.
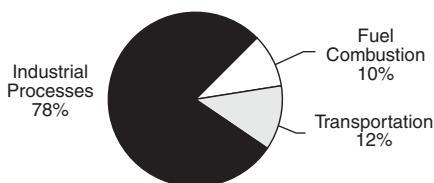
Regional Trends

Figure 2-13 segregates the ambient trend analysis by EPA Region. Although most Regions showed large concentration reductions between 1982 and 2001, there were some intermittent upturns, including a rather large upturn in the Region 1 trends plot. Most of these "bumps" in the trends graphs can be attributed to the inherent variability and noise associated with data reported near minimum detectable levels.

2001/2002 Air Quality Status

The large reductions in long-term Pb emissions from transportation sources have changed the nature of the ambient Pb problem in the United States. Because industrial processes are now responsible for all violations of the Pb standard, the Pb monitoring strategy currently focuses on emissions from these point sources.

The map in Figure 2-14 shows the highest quarterly mean Pb concentration by county in 2001. One area, with a total population of 201,219, containing some of the point sources identified in Figure 2-14 did not meet the Pb NAAQS in 2001.

Figure 2-13. Trend in Pb maximum quarterly mean concentration by EPA Region, 1982–2001.
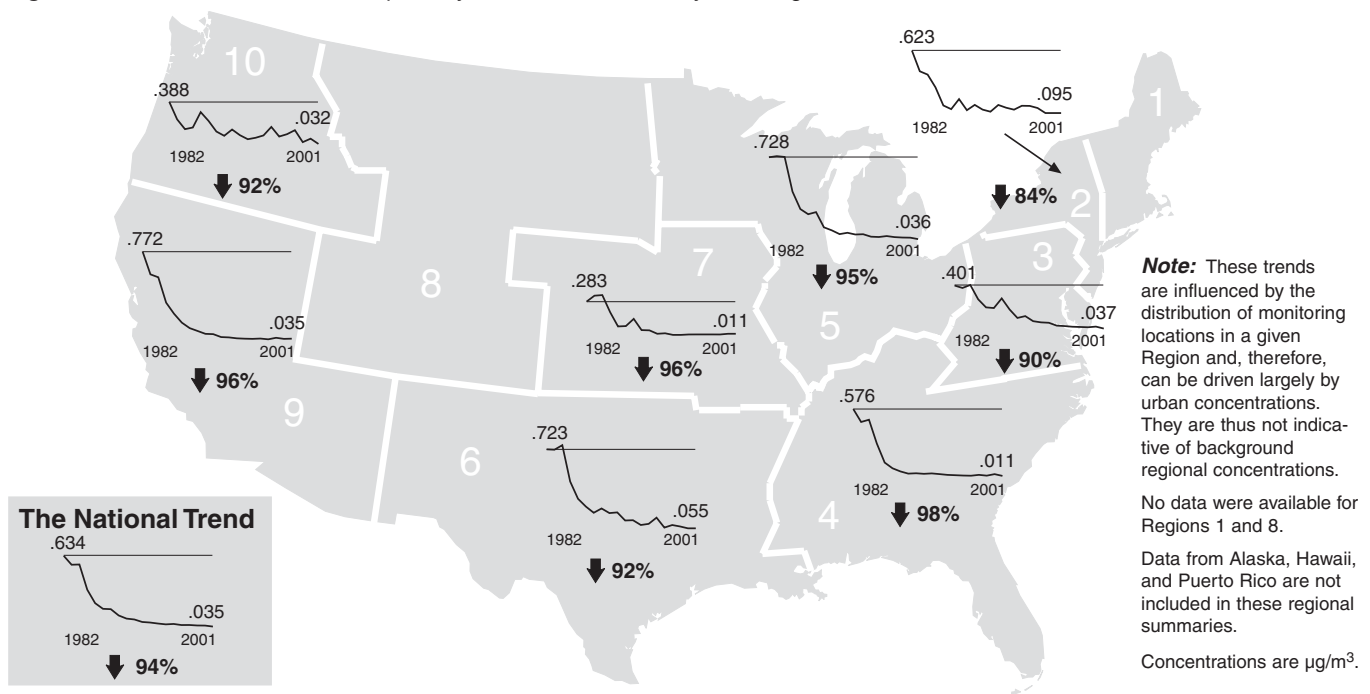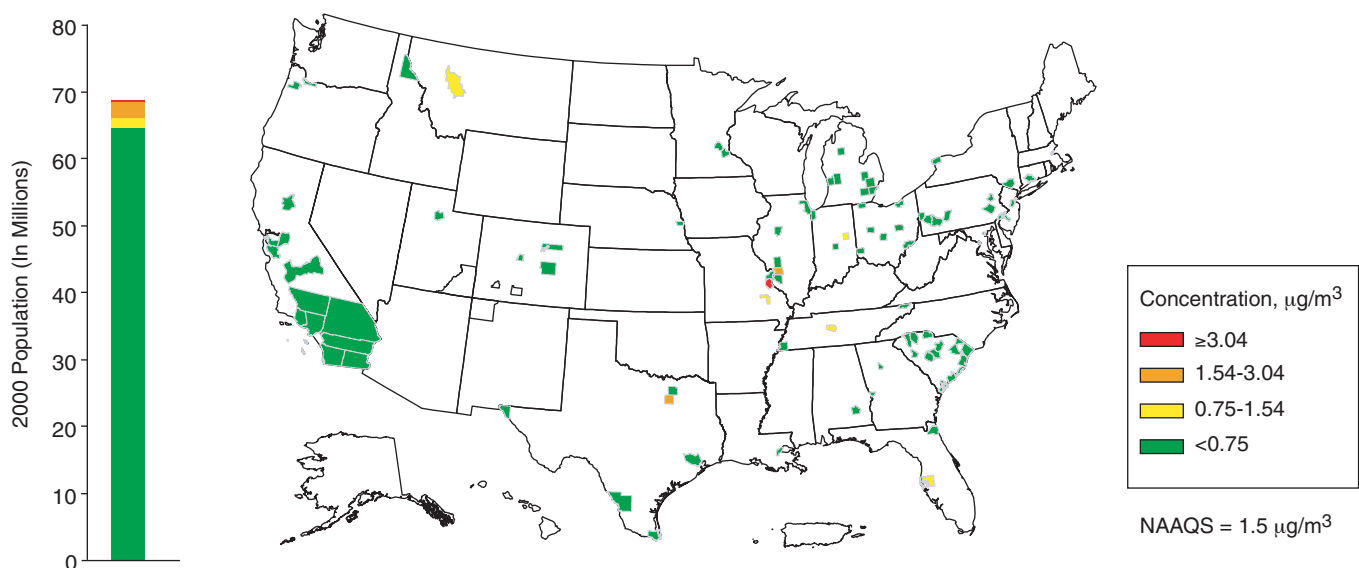

The National Trend

Note: These trends are influenced by the distribution of monitoring locations in a given Region and, therefore, can be driven largely by urban concentrations. They are thus not indicative of background regional concentrations.

No data were available for Regions 1 and 8.

Data from Alaska, Hawaii, and Puerto Rico are not included in these regional summaries.

Concentrations are µg/m^3.

Figure 2-14. Highest Pb maximum quarterly mean by county, 2001.

Concentration, µg/m^3

- ≥3.04
- 1.54-3.04
- 0.75-1.54
- <0.75

NAAQS = 1.5 µg/m^3

Nitrogen Dioxide

Air Quality Concentrations		
1983–02	21%	decrease
1993–02	11%	decrease

Emissions		
1983–02	15%	decrease
1993–02	12%	decrease

Worth Noting

- Over the past 20 years, nitrogen dioxide (NO_2) concentrations across the country have decreased significantly.

- All areas of the country that once violated the national air quality standard for NO_2 now meet that standard.

Nature and Sources

Nitrogen dioxide is a reddish-brown, highly reactive gas that is formed in the ambient air through the oxidation of nitric oxide (NO). Nitrogen oxides (NO_x), the term used to describe the sum of NO, NO_2, and other oxides of nitrogen, play a major role in the formation of ozone in the atmosphere through a complex series of reactions with VOCs. A variety of NO_x compounds and their transformation products occur both naturally and as a result of human activities. Anthropogenic (i.e., man-made) emissions of NO_x account for a large majority of all nitrogen inputs to the environment. The major sources of anthropogenic NO_x emissions are high-temperature combustion processes, such as those occurring in automobiles and power plants. Most NO_x from combustion sources (about 95 percent) are emitted as NO; the remainder are largely NO_2. Because NO is readily converted to NO_2 in the environment, the emissions estimates reported here assume nitrogen oxides are in the NO_2 form. Natural sources of NO_x are lightning, biological and abiological processes in soil, and stratospheric intrusion. Ammonia and other nitrogen compounds produced naturally are important in the cycling of nitrogen through the ecosystem. Home heaters and gas stoves also produce substantial amounts of NO_2 in indoor settings.

Health and Environmental Effects

Nitrogen dioxide is the most widespread and commonly found nitrogen oxide and is a matter of public health concern. The most troubling health effects associated with short-term exposures (i.e., less than 3 hours) to NO_2 at or near the ambient NO_2 concentrations seen in the United States include cough and increased changes in airway responsiveness and pulmonary function in individuals with preexisting respiratory illnesses, as well as increases in respiratory illnesses in children 5 to 12 years old.[5,6] Evidence suggests that long-term exposures to NO_2 may lead to increased susceptibility to respiratory infection and may cause structural alterations in the lungs.

Atmospheric transformation of NO_x can lead to the formation of ozone and nitrogen-bearing particles (e.g., nitrates and nitric acid). As discussed in the ozone and particulate matter sections of this chapter, exposure to both PM and O_3 is associated with adverse health effects.

Nitrogen oxides contribute to a wide range of effects on public welfare and the environment, including global warming and stratospheric ozone depletion. Deposition of nitrogen can lead to fertilization, eutrophication, or acidification of terrestrial, wetland, and aquatic (e.g., fresh water bodies, estuaries, and coastal water) systems. These effects can alter competition between existing species, leading to changes in the number and type of species (composition) within a community. For example, eutrophic conditions in aquatic systems can produce explosive algae growth leading to a depletion of oxygen in the water and/or an increase in levels of toxins harmful to fish and other aquatic life.

Primary and Secondary Standards

The level for both the primary and secondary NAAQS for NO_2 is 0.053 ppm annual arithmetic average (mean), not to be exceeded. In this report, the annual arithmetic average (mean) concentration is the metric used to evaluate and track ambient NO_2 air quality trends.

National Air Quality Trends

Since 1983, monitored levels of NO_2 have decreased 21 percent.[7] These downward trends in national NO_2 levels are reflected in all regions of the country. Nationally, average NO_2 concentrations are well below the NAAQS and are currently at the lowest levels recorded in the past 20 years. All areas of the country that once violated the NAAQS for NO_2 now meet that standard. Over the past 20 years, national emissions of NO_x have declined by almost 15 percent. Annual mean NO_2 concentrations declined in the early 1980s, were relatively unchanged during the mid-to-late 1980s, and resumed their decline in the 1990s. Figure 2-15 shows that the national composite annual mean NO_2 concentration in 2002 is 11 percent lower than that recorded in 1993. Except for 1994 and 1999, NO_2 concentrations have decreased, or remained unchanged, each year since 1989.

Figure 2-16 reveals how the trends in annual mean NO_2 concentrations vary among rural, suburban, and urban locations. The highest annual mean NO_2 concentrations are typically found in urban areas, with significantly lower annual mean concentrations recorded at rural sites.

Interestingly, as the nation has experienced these significant decreases in NO_2 concentrations, NO_x emissions are increasing, as described in more detail later in this section of the chapter. One possible explanation involves the location of the majority of the nation's NO_2 monitors. Most NO_2 monitoring sites are mobile-source-oriented sites in urban areas, and the 20-year decline in ambient NO_2 levels closely tracks the 19 percent reduction in emissions from gasoline-powered vehicles over the same time period.

Regional Air Quality Trends

The map in Figure 2-17 provides regional trends in NO_2 concentrations during the past 20 years, 1982 to 2001 (except Region 10, which does not have any NO_2 trend sites). The trends seen in the suburban and urban sites track the declining trend in NO_x emissions, as compared with the trend in rural sites. The trends statistic is the regional composite mean of the NO_2 annual mean concentrations across all sites with at least 8 years of ambient measurements. The largest reductions in NO_2 concentrations occurred in the south coast of California and parts of the Northeast and Mid-Atlantic states. Slightly smaller reductions in mean NO_2 concentrations were recorded in New England, the Southeast, and the Southwest. Interestingly, NO_2 concentrations were unchanged in the Midwest states and have actually increased in the North Central states.

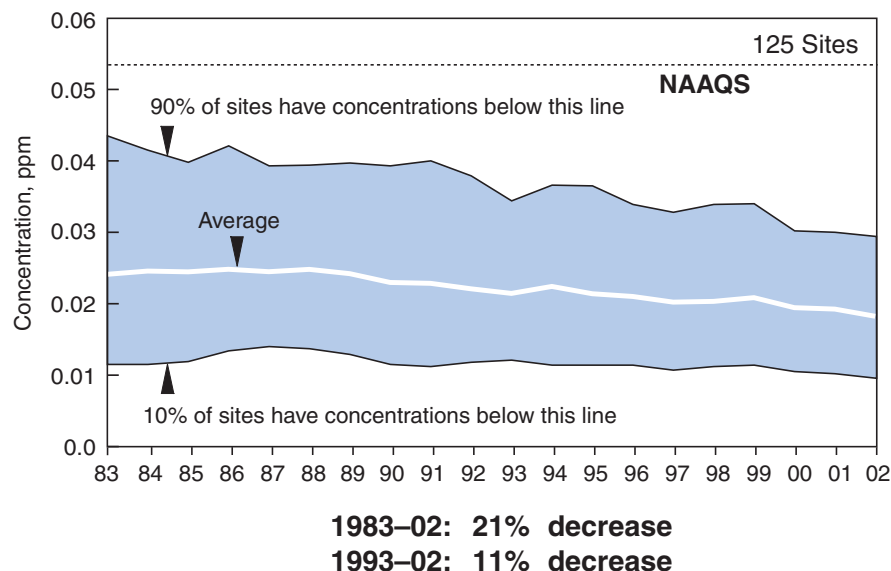**Figure 2-15**. NO_2 air quality, 1982–2001, based on annual arithmetic average.

1983–02: 21% decrease
1993–02: 11% decrease

Figure 2-16. Trend in annual mean NO_2 concentrations by type of location, 1982–2001.

|	1982-2001
Rural Sites	14
Suburban Sites	58
Urban Sites	49
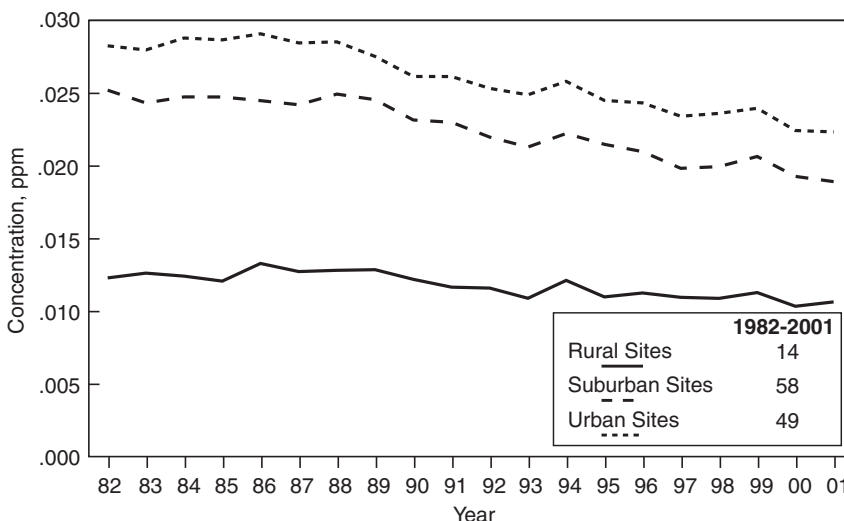
Figure 2-17. Trend in NO₂ maximum quarterly mean concentration by EPA Region, 1982–2001.

Note: These trends are influenced by the distribution of monitoring locations in a given Region and, therefore, can be driven largely by urban concentrations. They are thus not indicative of background regional concentrations.

Data from Alaska, Hawaii, and Puerto Rico are not included in these regional summaries.

Concentrations are ppm.
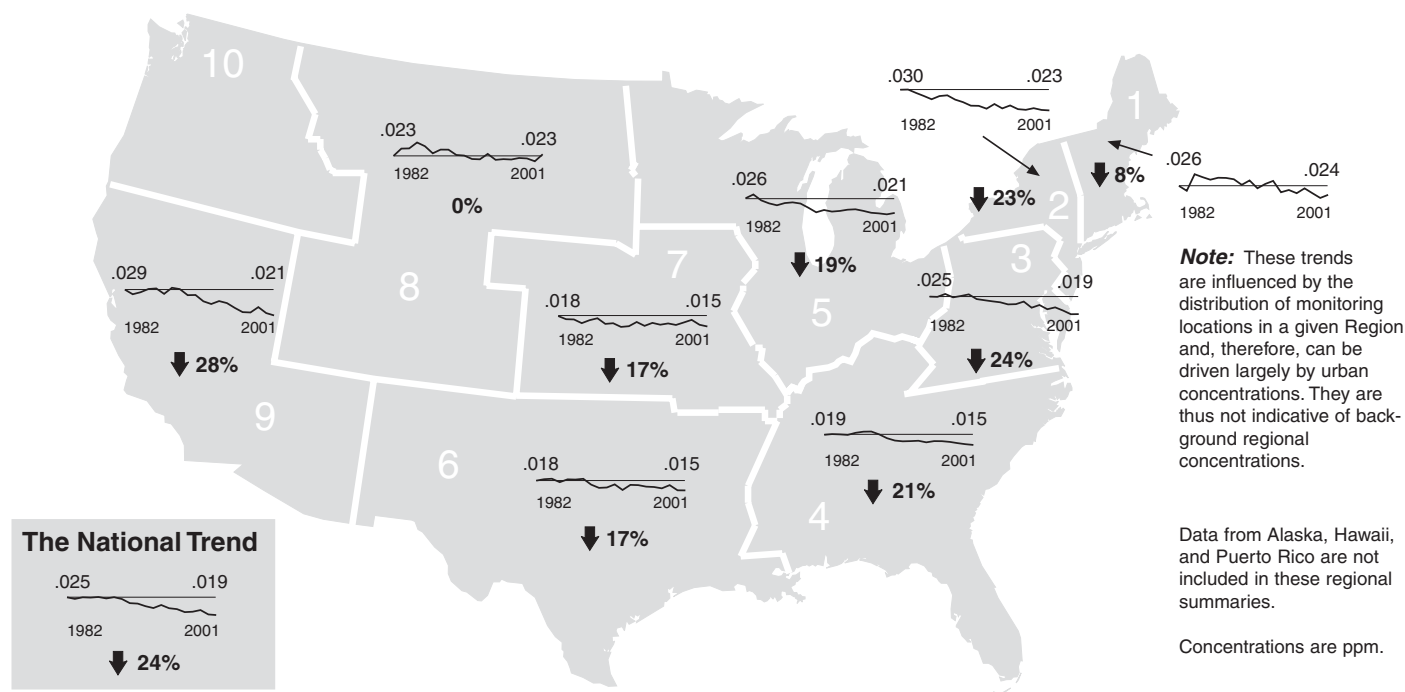
This increase coincides with increases in NO$_x$ emissions from transportation (both onroad and nonroad) as well as power plants in selected states with NO$_2$ monitors in these areas.

National Emissions Trends

The reduction in emissions for NO$_x$ shown in Figure 2-18 differs from the increase in NO$_x$ emissions reported in previous editions of this report. These emission trends reflect new and improved emission estimates for highway vehicles and nonroad engines. While NO$_x$ emissions are declining overall, emissions from some sources such as nonroad engines have actually increased since 1983. These increases are of concern given the significant role NO$_x$ emissions play in the formation of ground-level ozone (smog) as well as other environmental problems like acid rain and nitrogen loadings to

Figure 2-18. NO$_x$ emissions, 1983–2002.
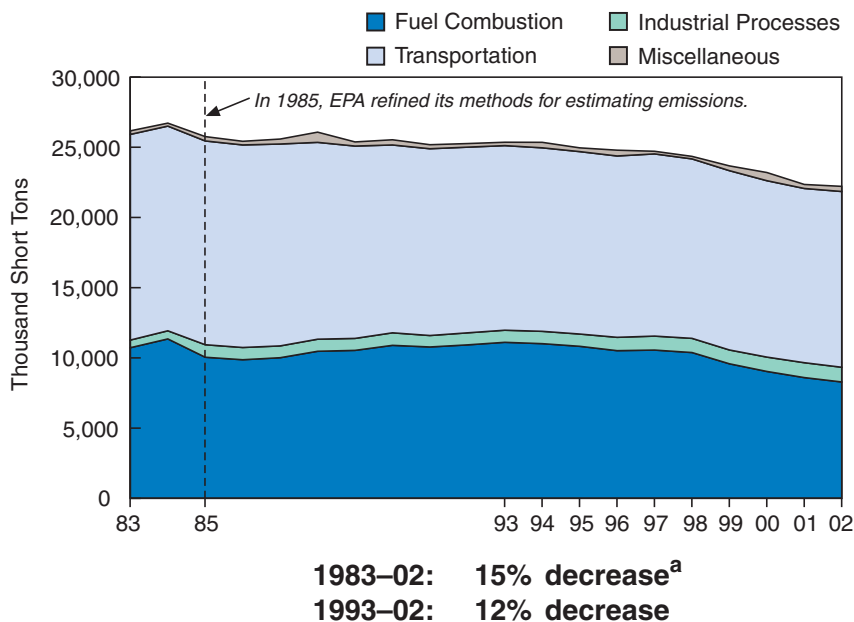
1983–02: 15% decrease[a]
1993–02: 12% decrease

Note: Emission estimation methods and data sources have evolved over time, resulting in some inconsistency in estimates in different years. In the methods used for this report, the significant changes have occurred between 1984 and 1986, and between 1995 and 1996, although not all source types were affected. More explanation is provided in Appendix B.

[a] Emissions trends data are not available for 1983; thus, the 20-year trend was interpolated based on emissions data for 1980 and 1985.

waterbodies described above. In response, EPA has proposed regulations that will significantly control NO_x emissions from nonroad diesel engines.

Figure 2-19 indicates that the two primary sources of NO_x emissions are transportation and stationary source fuel combustion. Together, these two sources make up 93 percent of 2002 total NO_x emissions. Emissions from transportation sources have decreased 15 percent over the past 20 years and decreased 5 percent during the past 10 years. For both light-duty gasoline vehicles and light-duty gasoline trucks, NO_x emissions peaked in 1994 and then began a steady decrease through 2000. This decrease can be attributed primarily to the implementation of the Tier 1 emission standards that lowered NO_x emissions from new cars and light-duty trucks. In contrast, NO_x emissions from heavy-duty vehicles, both gasoline and diesel, decreased significantly over the 10-year period (17 percent

decrease for gasoline and 12 percent increase for diesel). A portion of this increase is due to the increase in VMT for these categories for heavy-duty gasoline vehicles and diesel trucks. In addition, emissions from heavy-duty diesel vehicles increased over this period due to the identification of "excess emissions" in many diesel vehicles. These excess emissions peaked in 1998, and emissions of heavy-duty diesel vehicles are now declining. New emission standards will lead to further reductions in emissions from heavy duty vehicles in the future. Further, emissions from nonroad vehicles, particularly those fueled with diesel, have steadily increased over the last 10 years. EPA is developing new standards to reduce these emissions.

Reductions in NO_x emissions from fuel combustion, particularly those from electric power generator units in the past 2 years, have partially offset the impact of increases in the transportation sector. Emissions from these generator units in 2001

were 5 percent lower than they were in 2000. The Acid Deposition Control provisions of the Act (Title IV) required EPA to establish NO_x annual emission limits for coal-fired electric utility units in two phases, resulting in NO_x reductions of approximately 400,000 tons per year during Phase I (1996–1999) and 2 million tons per year in Phase II (year 2000 and subsequent years).[8]

Figure 2-20 shows the geographic distribution of 2001 NO_x emissions based on the tonnage per square mile for each county. This map illustrates that the eastern half of the country and the West Coast emit more NO_x (on a density basis) than does the western half of the continental United States.

2001 Air Quality Status

All monitoring locations across the nation met the NO_2 NAAQS in 2001. This is reflected in Figure 2-21, which displays the highest annual mean NO_2 concentration measured in each county.

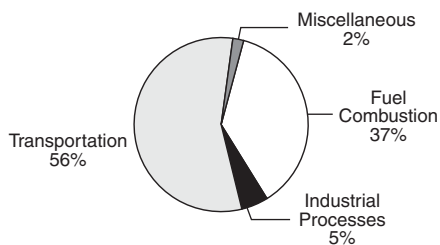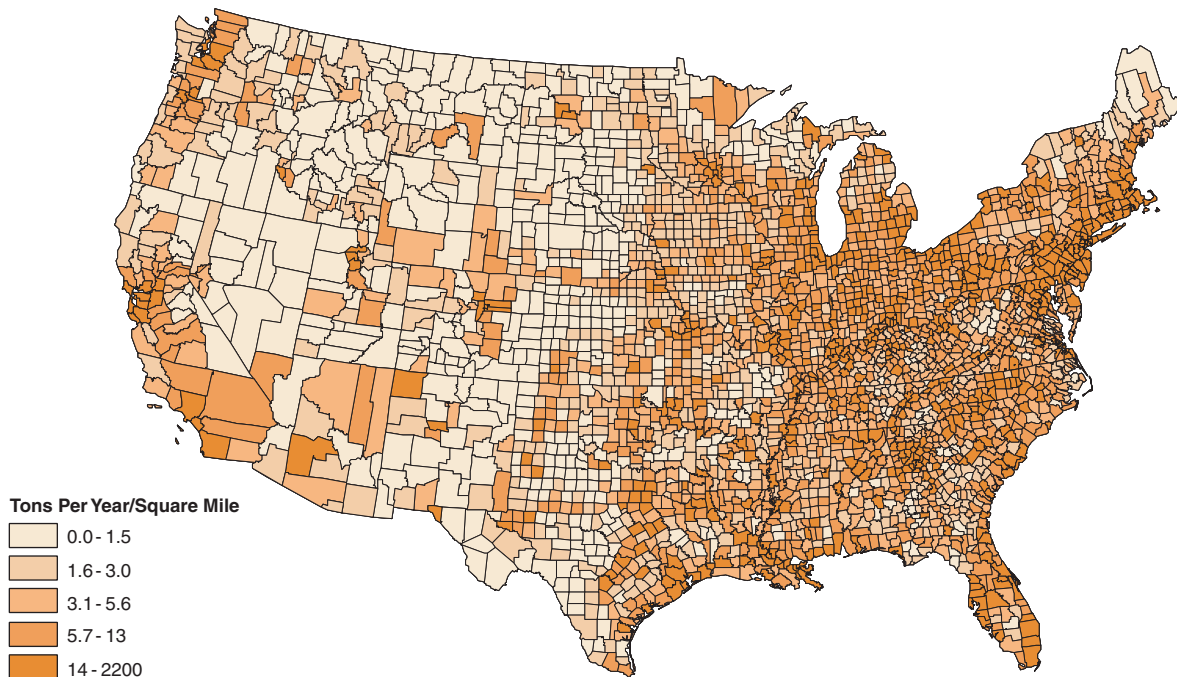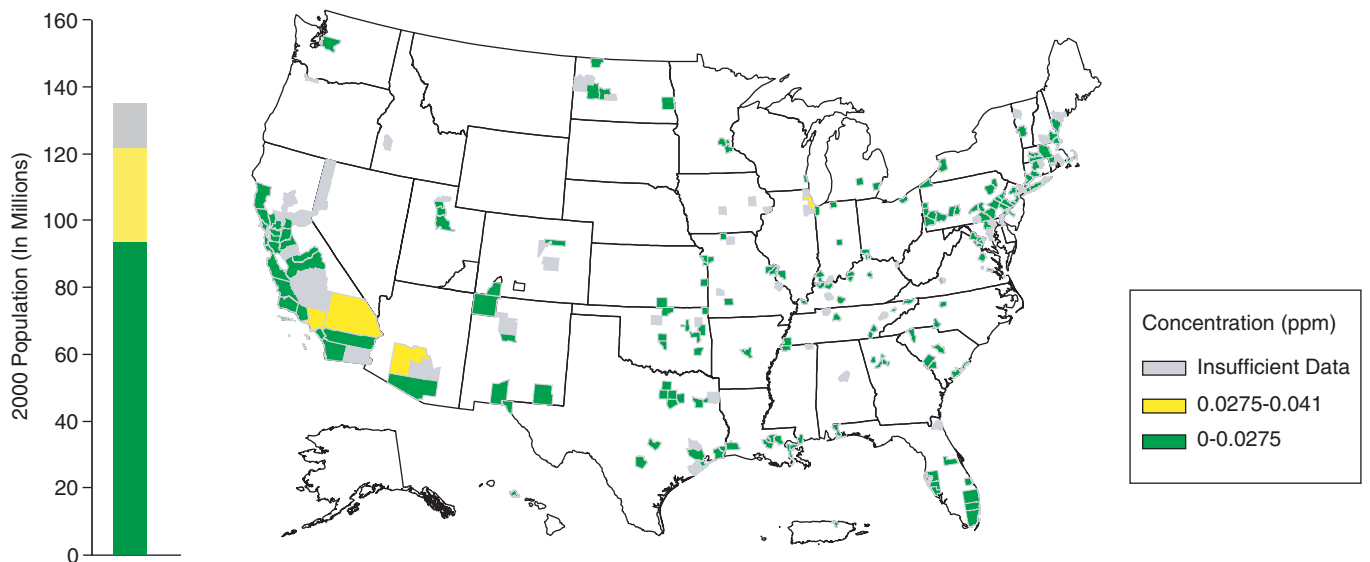**Figure 2-19**. NO_x emissions by source category, 2002.

Figure 2-20. Density map of 2001 NO$_2$ emissions, by county.

Tons Per Year/Square Mile
- 0.0 - 1.5
- 1.6 - 3.0
- 3.1 - 5.6
- 5.7 - 13
- 14 - 2200

Figure 2-21. Highest NO$_2$ maximum quarterly mean by county, 2001.

Concentration (ppm)
- Insufficient Data
- 0.0275-0.041
- 0-0.0275

Ozone

Air Quality Concentrations		
1983–02	22%	decrease (1-hr)
	14%	decrease (8-hr)
1993–02	2%	decrease (1-hr)
	4%	increase (8-hr)

Emissions (Anthropogenic VOCs)	
1983–02	40% decrease
1993–02	25% decrease

Worth Noting

- Over the past 20 years, ozone (O_3) levels (1-hour and 8-hour) have improved considerably nationwide.

- However, over the past 10 years, ozone levels (1-hour and 8-hour) have been relatively flat.

Nature and Sources

Ground-level O_3 remains a pervasive pollution problem in the United States. Ozone is readily formed in the atmosphere by the reaction of VOCs and NO_x in the presence of heat and sunlight, which are most abundant in the summer. VOCs are emitted from a variety of sources, including motor vehicles, chemical plants, refineries, factories, consumer and commercial products, other industries, and natural (biogenic) sources. Nitrogen oxides (a precursor to ozone) are emitted from motor vehicles, power plants, and other sources of combustion, as well as natural sources including lightning and biological processes in soil. Changing weather patterns contribute to yearly differences in O_3 concentrations. Ozone and the precursor pollutants that cause O_3 also can be transported into an area from pollution sources located hundreds of miles upwind.

Health and Environmental Effects

Ozone occurs naturally in the stratosphere and provides a protective layer high above the Earth. However, at ground level, it is the prime ingredient of smog. Short-term (1- to 3-hour) and prolonged (6- to 8-hour) exposures to ambient O_3 concentrations have been linked to a number of health effects of concern. For example, increased hospital admissions and emergency room visits for respiratory causes have been associated with ambient O_3 exposures.

Exposures to O_3 result in lung inflammation, aggravate preexisting respiratory diseases such as asthma, and may make people more susceptible to respiratory infection. Other health effects attributed to short-term and prolonged exposures to O_3, generally while individuals are engaged in moderate or heavy exertion, include significant decreases in lung function and increased respiratory symptoms such as chest pain and cough. Children active outdoors during the summer when O_3 levels are at their highest are most at risk of experiencing such effects. Other at-risk groups include adults who are active outdoors, such as outdoor workers, and individuals with preexisting respiratory disorders such as asthma and chronic obstructive lung disease. Within each of these groups are individuals who are unusually sensitive to O_3. In addition, repeated long-term exposure to O_3 presents the possibility of irreversible changes in the lungs, which could lead to premature aging of the lungs and/or chronic respiratory illnesses.

Ozone also affects sensitive vegetation and ecosystems. Specifically, O_3 can lead to reductions in agricultural and commercial forest yields, reduced survivability of sensitive tree seedlings, and increased plant susceptibility to disease, pests, and other environmental stresses such as harsh weather. In long-lived species, these effects may become evident only after several years or even decades. As these species are out-competed by others, long-term effects on forest ecosystems and habitat quality for wildlife and endangered species become evident. Furthermore, O_3 injury to the foliage of trees and other plants can decrease the aesthetic value of ornamental species as well as the natural beauty of our national parks and recreation areas.

Primary and Secondary 1-hour Ozone Standards

In 1979, EPA established 1-hour primary and secondary standards for O_3. The level of the 1-hour primary and secondary O_3 NAAQS is 0.12 ppm daily maximum 1-hour concentration that is not to be exceeded more than once per year on average.

Primary and Secondary 8-hour Ozone Standards

On July 18, 1997, EPA strengthened the O_3 NAAQS based on the latest scientific information showing adverse effects from exposures allowed by the then-existing standards. The standard was set in terms of an 8-hour averaging time.[9]

Refer to **http://www.epa.gov/airlinks** for up-to-date information concerning actions surrounding the revised standards.

Air Quality Trends

Because the 1-hour and 8-hour NAAQS have different averaging times and forms, two different statistics are used in this report to track

ambient O_3 air quality trends. For the 1-hour O_3 NAAQS, this report uses the composite mean of the annual second-highest daily maximum 1-hour O_3 concentration as the statistic to evaluate trends. For the 8-hour O_3 NAAQS, this report relies on the annual fourth-highest 8-hour daily maximum O_3 concentration as the statistic of interest to assess trends.

National Air Quality Trends

Figure 2-22 clearly shows that, over the past 20 years, peak 1-hour O_3 concentrations have declined considerably at monitoring sites across the country. From 1983 to 2002, national 1-hour O_3 levels improved 22 percent, with 1983, 1988, and 1995 representing peak years for this pollutant. Figure 2-22 shows that 370 sites met the data completeness criteria over the past 20 years (1983–2002). It is important to interpret such long-term, quantitative ambient O_3 trends carefully given changes in network design, siting criteria, spatial coverage, and monitoring instrument calibration procedures during the past two decades. More recently, national 1-hour O_3 levels have continued to improve, but the progress has been less rapid, as evidenced by the 2 percent decrease from 1993 to 2002.

Figure 2-23 shows the national trend in 8-hour O_3 concentrations across the same sites used to estimate the national 1-hour O_3 trends. Nationally, 8-hour levels have decreased 14 percent over the last 20 years. However, just as is true for the 1-hour levels, the progress in 8-hour O_3 levels over the last 10 years has slowed and actually shows a 4 percent increase in national levels between 1993 and 2002. Standard statistical tests applied to the 10-year trends for both 1-hour and 8-hour ozone shows that these trends are not statistically significant. Ozone

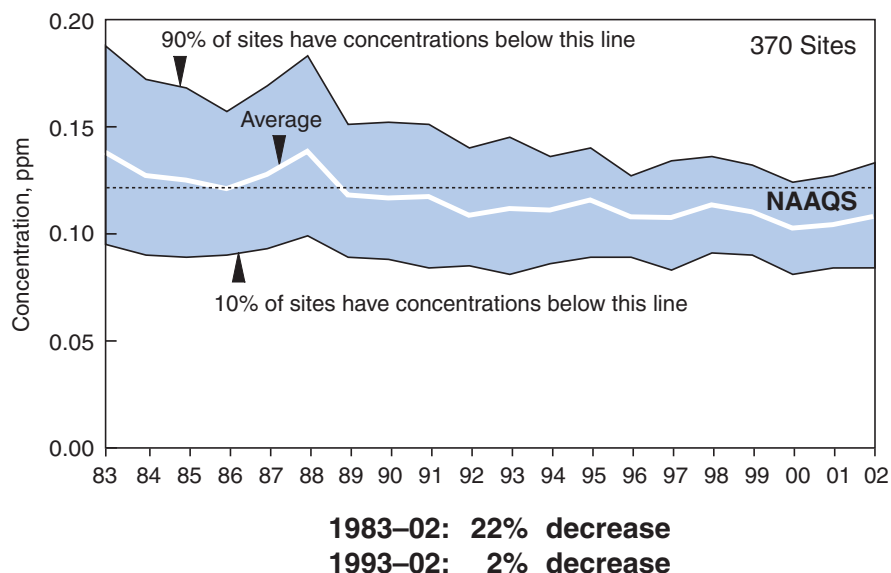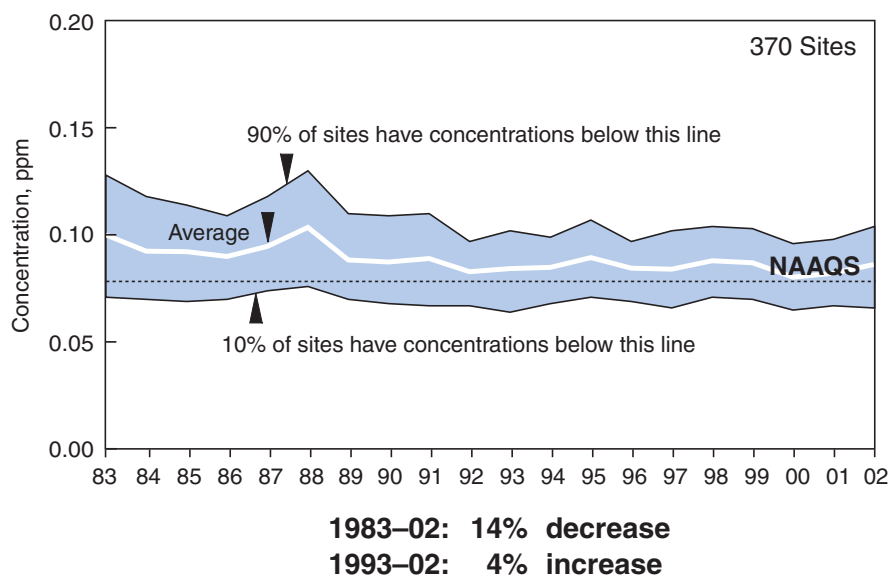**Figure 2-22.** O_3 air quality, 1983–2002, based on annual second maximum 1-hour average.

1983–02: 22% decrease
1993–02: 2% decrease

Figure 2-23. O_3 air quality, 1983–2002, based on annual fourth maximum 8-hour average.

1983–02: 14% decrease
1993–02: 4% increase

concentrations varied over this 10-year period from year to year but did not change overall. The trend in the 8-hour O_3 statistic is similar to the trend in the 1-hour values, although the concentration range is smaller.

Regional Air Quality Trends
The map in Figure 2-24 examines trends in 1-hour O_3 concentrations during the past 20 years by geographic region of the country. The 1-hour O_3 levels in all areas of the country have generally followed the pattern of declining trends since 1982 similar to that of the national observations. However, the magnitude of improvement has not been consistent across all regions.

Figure 2-24. Trend in 1-hour O_3 levels, 1983–2002, averaged across EPA Regions, based on annual second highest daily maximum.
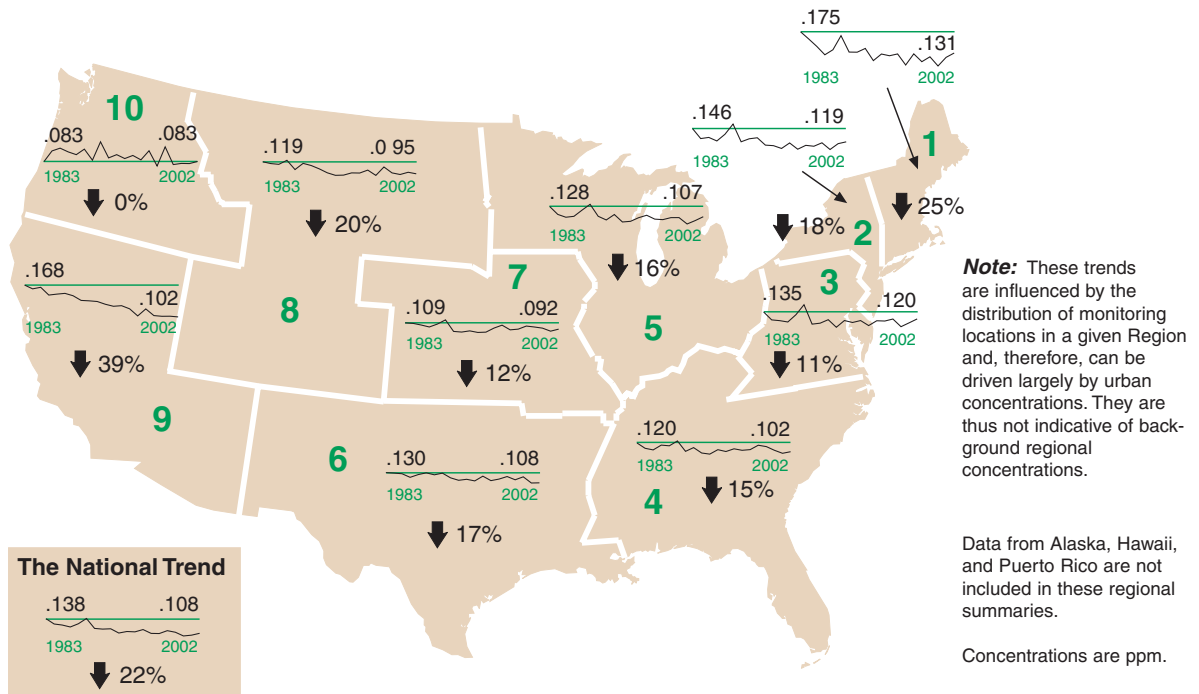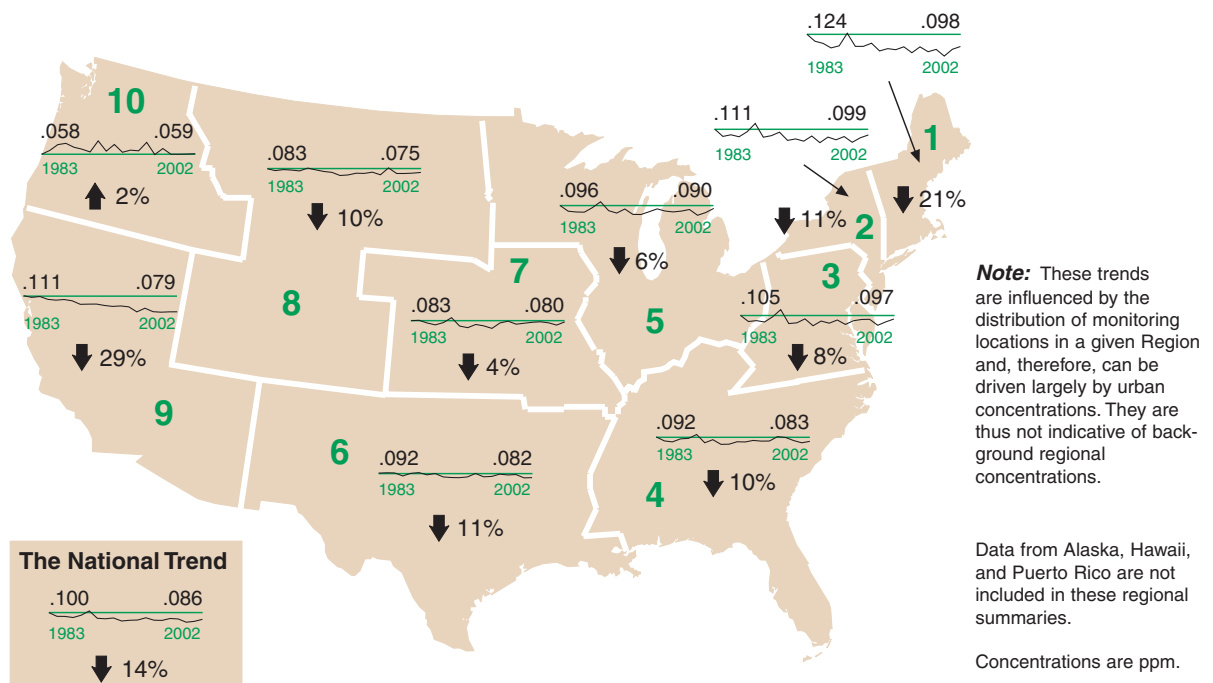

Note: These trends are influenced by the distribution of monitoring locations in a given Region and, therefore, can be driven largely by urban concentrations. They are thus not indicative of background regional concentrations.

Data from Alaska, Hawaii, and Puerto Rico are not included in these regional summaries.

Concentrations are ppm.

Figure 2-25. Trend in 8-hour O_3 levels, 1983–2002, averaged across EPA Regions, based on annual fourth maximum 8-hour average.

Note: These trends are influenced by the distribution of monitoring locations in a given Region and, therefore, can be driven largely by urban concentrations. They are thus not indicative of background regional concentrations.

Data from Alaska, Hawaii, and Puerto Rico are not included in these regional summaries.

Concentrations are ppm.

Similarly, Figure 2-25 portrays 8-hour O_3 trends by geographic region of the country. Again, most areas of the country show 20-year air quality improvements (with respect to 8-hour O_3) consistent with the national trend, with the most significant improvements occurring in the Northeast and Pacific Southwest. The Pacific Northwest region showed a slight increase in the 8-hour ozone for the period 1983–2002.

In Figure 2-26, the national 1-hour O_3 trend is disaggregated to show the 20-year change in ambient O_3 concentrations among rural, suburban, and urban monitoring sites. The highest ambient O_3 concentrations are typically found at suburban sites, consistent with the downwind transport of emissions from the urban center. During the past 20 years, O_3 concentrations decreased by approximately 23 percent at suburban sites, and 26 percent at urban sites. At rural sites, 1-hour O_3 levels for 2002 are approximately 16 percent lower than they were in 1983 and, for the sixth consecutive year, are greater than the level observed for urban sites.

Urban Area Air Quality Trends

It is important to note that year-to-year changes in ambient ozone trends are influenced by meteorological conditions, population growth, and changes in emission levels of ozone precursors (i.e., VOCs and NO_x) resulting from ongoing control measures. For example, to further evaluate the 10-year 8-hour ozone trends, EPA applied a model to the annual rate of change in ozone based on measurements in 53 metropolitan areas (Figure 2-27). This model adjusted the ozone data in these areas to account for the influence of local meteorological conditions, including surface temperature and

windspeed. Figure 2-27 shows the aggregated trend in 8-hour ozone for these 53 areas adjusted for meteorological conditions for the 10-year period 1993-2002. The figure also shows the aggregated trend for these areas unadjusted for meteorology and the national average in 8-hour ozone. From this figure, the

meteorologically adjusted trend for this 10-year period can be seen as relatively flat.

EPA's analysis of ambient ozone concentration data indicates that ozone concentrations are on the increase in some urban areas. These increases are evident based on both 1-hour and 8-hour trends, as shown

Figure 2-26. Trend in annual second-highest daily maximum 1-hour O_3 concentrations by location, 1983–2002.
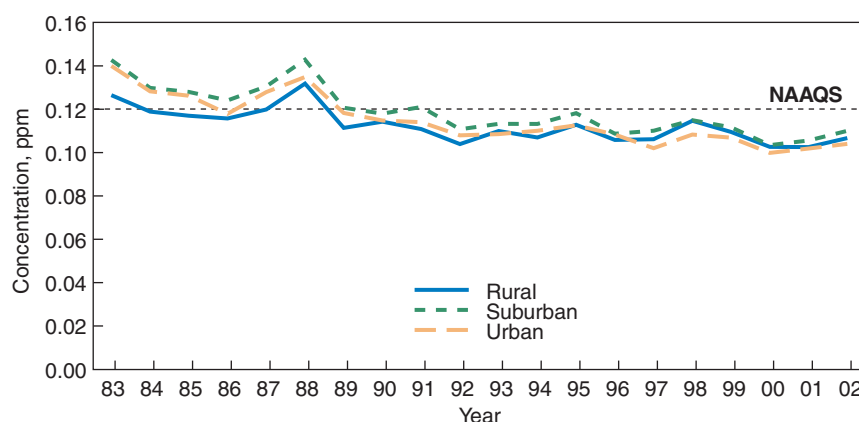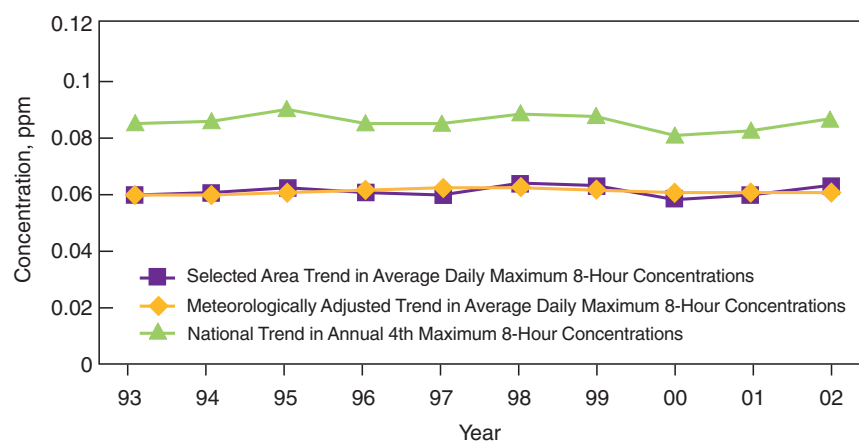

Figure 2-27. Comparison of actual and meteorologically adjusted 8-hour O_3 trends, 1993–2002.

in Figures 2-28 and 2-29. Ozone concentrations are on the increase in several cities in the southeastern and midwestern United States, while urban areas on the West Coast and in New England generally show decreasing trends. Figures 2-28 and 2-29 show a comparison of ozone trends over two consecutive 10-year time frames. The 1-hour trends show an increasing number of cities with upward ozone trends in the western

**Figure 2-28**. 1-Hour O$_3$ trends for 1991–2000 and 1992–2001.
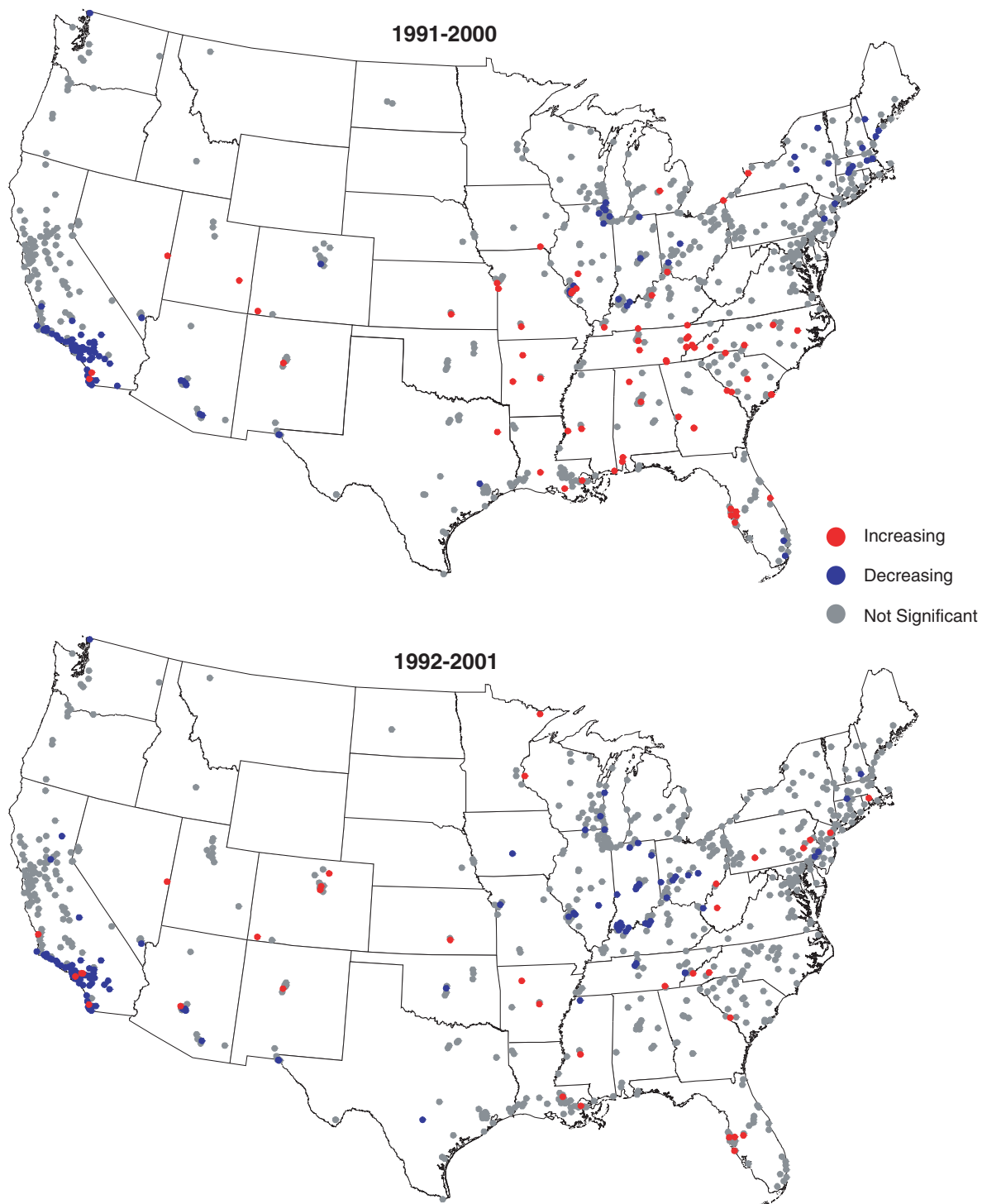
and mid-Atlantic urban areas and a decreasing number of cities with upward ozone trends in the Southeast. The 8-hour ozone trends also show a decrease in the number of

cities with upward ozone trends in the Southeast, but an increasing number of cities with upward trends in New England and around the Great Lakes.

Trends at PAMS Sites

Photochemical Assessment Monitoring Program Stations (PAMS) are operated by states in areas that were originally classified as extreme,

Figure 2-29. 8-Hour O$_3$ trends for 1991–2000 and 1992–2001.
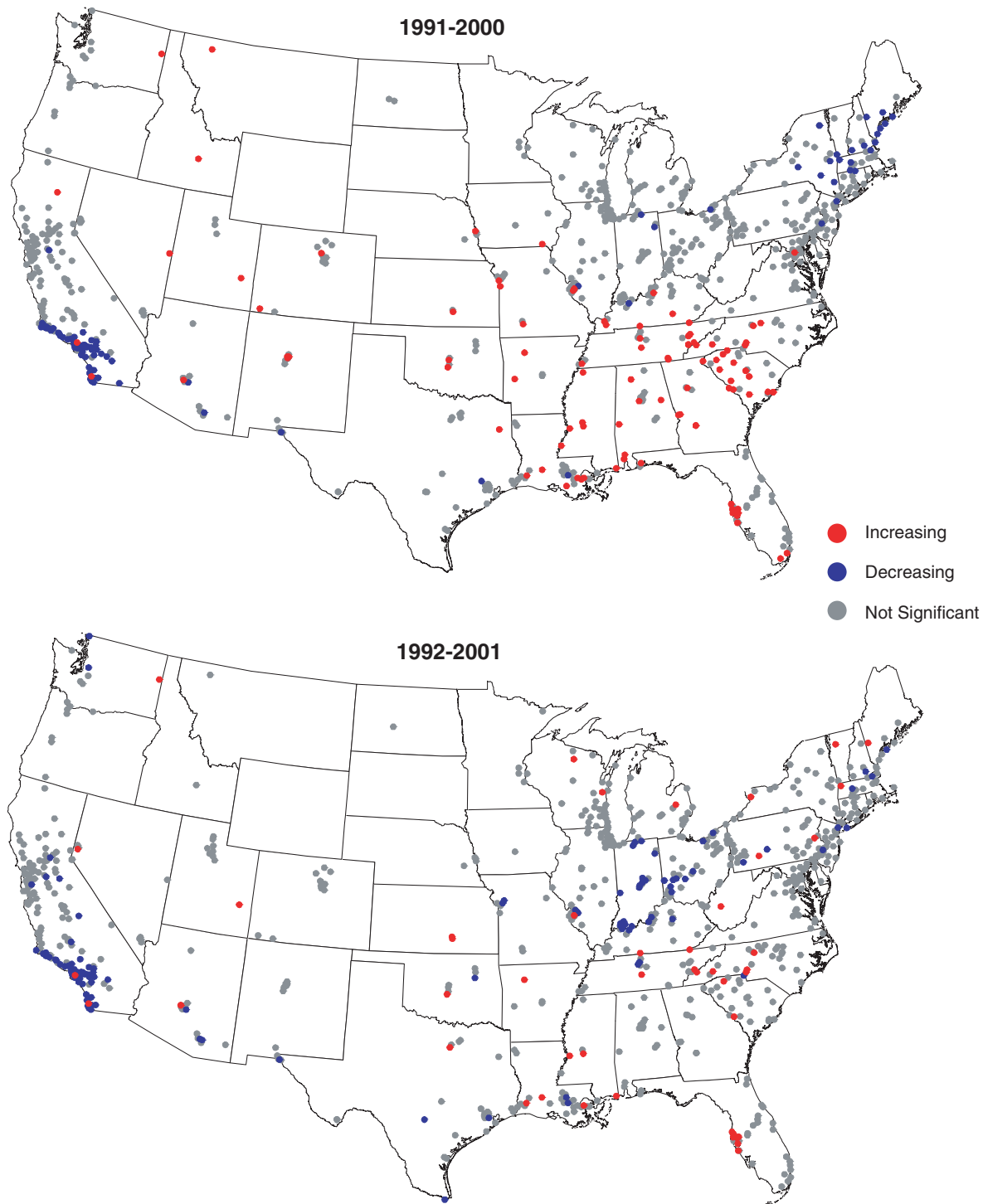
severe, or serious nonattainment for ozone. Ozone, ozone precursor, and surface and upper air meteorological conditions are monitored at PAMS sites during the summer months when meteorological conditions are most conducive to ozone formation. Some PAMS sites have been in operation since 1994 and there are now sufficient data available to examine long-term air quality trends. Trends in total nonmethane organic compounds (TNMOC), NO_x, and selected VOC species at PAMS locations are tabulated in Table 2-3; median percent changes are illustrated in Figure 2-30. These trends are for concentrations averaged over the hours from 6 to 9 a.m. when ozone precursor concentrations are typically at their maximum and best represent the influence of fresh, local emissions. VOC species were selected for inclusion in this analysis based primarily on relative abundance and status as a hazardous air pollutant under the Clean Air Act. Trends in other VOC species monitored under the PAMS program can generally be expected to be similar to those shown here.

All species except isoprene and NO_x exhibited substantial median percentage declines over the 1995 to 2001 trend period. Isoprene is largely emitted by biogenic sources (trees and other vegetation) and would therefore not be expected to show a significant trend. For TNMOC and TNMOC species other than isoprene, concentrations decreased at all or nearly all sites, although the decline was not statistically significant in every case. NO_x concentrations increased at roughly one third of all sites, but none of these increases were found to be statistically significant. Trends at PAMS Type 2 sites, which are generally located within areas of maximum

**Table 2-3**. Trends in TNMOC, NO_x, and Selected VOC Species

| | All Site Types | | | | | Type 2 Sites | | | | | Median % Change | |
| | | All Sites[a] | | Stat. Significant[b] | | | All Sites[a] | | Stat. Significant[b] | | | |
	Total	Up	Down	Up	Down	Total	Up	Down	Up	Down	All Sites	Type 2 Sites
TNMOC	28	4	23	0	14	14	2	12	0	7	-32	-36
NO_x	63	16	33	0	7	25	6	16	0	2	-8	-9
Ethylene	21	1	15	0	7	12	0	10	0	4	-42	-40
Propylene	17	0	14	0	5	10	0	9	0	3	-40	-39
Isopentane	22	0	18	0	8	11	0	10	0	4	-30	-36
Isoprene	22	8	5	0	0	12	3	4	0	0	0	0
Benzene	22	0	18	0	12	12	0	12	0	9	-35	-43
Toluene	22	2	19	0	7	12	0	11	0	4	-29	-38
m/p-Xylene	22	0	20	0	9	12	0	12	0	6	-31	-33
o-Xylene	20	0	16	0	8	12	0	11	0	6	-36	-36
1,2,4-Trimethyl-benzene	20	4	12	0	4	11	0	9	0	4	-38	-57

TNMOC = Total nonmethane organic compound.

[a]Indicates sign of trend regardless of statistical significance.

[b]Indicates sign of trend at sites where trend is statistically significant at the 95% confidence level.

Notes:

1 The number of sites listed in the up and down columns indicates the number of PAMS locations at which the 1995–2001 trend in 6–9 a.m. average concentration is in the indicated direction. The number of sites in the total column may not equal the total of the up and down columns–either because the nonparametric trend estimate for some sites is identically zero or the trend at many sites is not statistically significant.

2 Theil's two-sided nonparametric significance test for the slope was used to assess statistical significance at the 95% confidence level consistent with the methodology used in previous National Air Quality and Emissions Trends reports. Note that these results are not adjusted for multiple comparisons.

Figure 2-30. Median percent change for the period 1995–2001 at PAMS monitors for selected species.
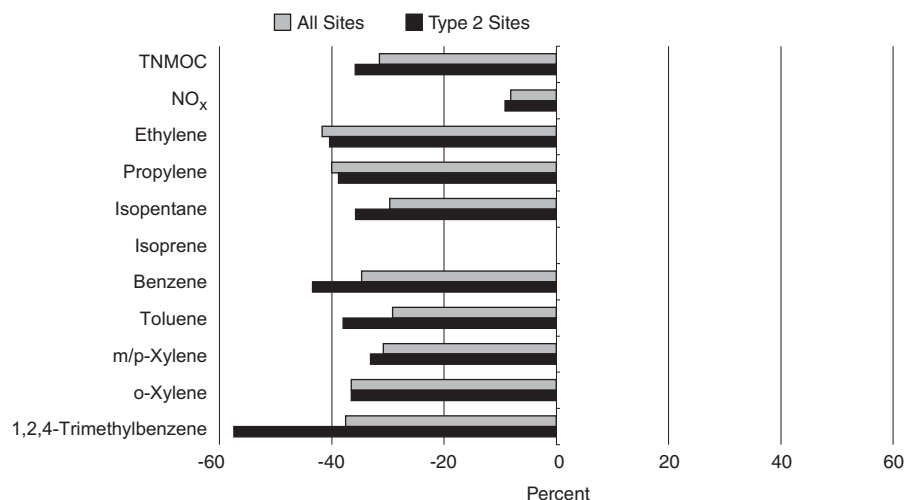
precursor emissions, are similar to trends over all site types although the Type 2 sites exhibited somewhat greater declines in isopentane, benzene, toluene, and 1,2,4-trimethyl-benzene.

Methodology

All data were obtained from EPA's Air Quality System (AQS) database. Trends are based on data from sites meeting certain data completeness criteria for the 1995–2001 period. Data completeness requirements are the same as those used in previous National Air Quality and Emissions Trends reports.[10] Annual averages computed from 1-hour samples of TNMOC or NO_x were considered valid if data were available for 50 percent or more of all possible observations. Sites selected for trends analysis must have valid annual summary statistics available for 5 or more years. Missing annual summary statistics were filled in via linear interpolation from surrounding years. If a missing value happened to fall at the beginning or end year of the period being investigated, the value was set equal to the nearest available valid year of data. Theil's nonparametric trend-slope estimates and two-sided significance test results for the slope were used to assess statistical significance consistent with the methodology used in previous National Air Quality and Emissions Trends reports. Note that these results are not adjusted for multiple comparisons.

Ozone and Ozone Precursor Trends in Chicago and Atlanta

Despite much progress in the years since passage of the 1990 Clean Air Act Amendments, some metropolitan areas are still classified as nonattainment with respect to the NAAQS

for 1-hour ozone. Two notable examples are Chicago and Atlanta. Atlanta is currently classified as a "serious" ozone nonattainment area; Chicago is currently classified as "severe." In this section we take a closer look at recent trends in ozone and ozone precursors in these two major metropolitan areas.

Composite ozone trends for 1-hour and 8-hour annual ozone design values in Chicago and Atlanta are depicted in Figure 2-31.[11] Trends in 1-hour design values are shown for the period 1991 to 2001; 8-hour design values are shown for the period 1996 to 2001 because 1996 is the first year for which EPA began reporting 8-hour design values. Design values vary from year to year, largely in response to changes in meteorological conditions that make it difficult to identify any long-term trend in either city.

Composite trends in summer weekday morning ozone precursor concentrations in Chicago and Atlanta are illustrated in Figure 2-32. Trends are shown for concentrations

on weekday mornings (6–9 a.m.), the period when precursor concentrations are typically at their maximum and are most directly influenced by fresh emissions from local sources. To maintain consistency between nonattainment areas and to retain sites in the analysis from Chicago that would otherwise not meet the data completeness criteria for a time period extending back to 1991, NO_x summary statistics were calculated for the period 1995 to 2001 only. TNMOC data are only available starting in 1995 for both nonattainment areas. An examination of Figure 2-34 indicates that TNMOC concentrations declined in both cities during this period, while NO_x concentrations increased slightly.

Air quality trend statistics for both cities are summarized in Table 2-4. Although none of the trends were found to be statistically significant, the results are generally consistent with a slight decrease in ozone accompanied by a more noticeable decrease in morning TNMOC and a slight increase in morning NO_x.

Figure 2-31. Annual 1-hour and 8-hour composite O_3 design values in the Atlanta and Chicago-Gary lake county nonattainment areas.
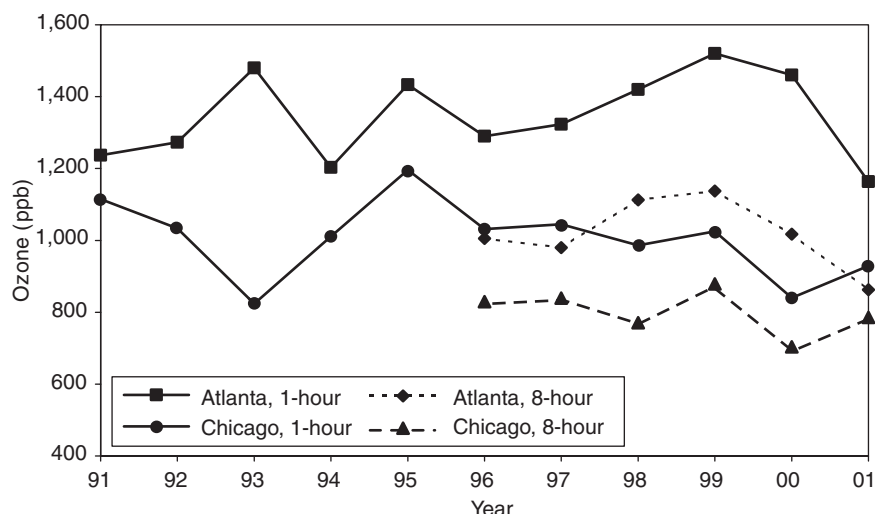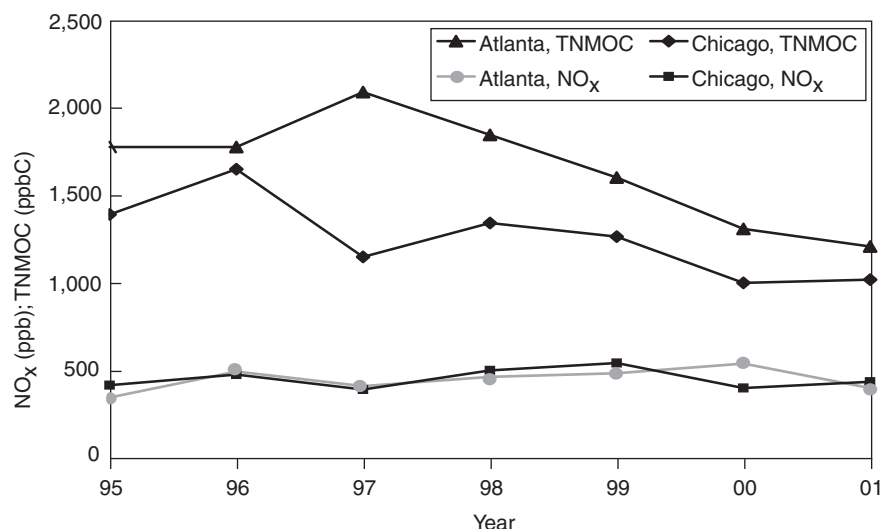
Figure 2-32. June-August weekday morning average NO_x and TNMOC at PAMS Type 2 trend sites (June 1–September 1, 6:00–9:00 a.m.).

Table 2-4. Summary of 1991–2001 Trends in Ozone Design Values and 1995–2001 Trends in Summer Weekday Morning Ozone Precursor Trends in Atlanta and Chicago

City	Pollutant	Composite Trend (ppb/year)	No. of Sites with Trend Increasing	No. of Sites with Trend Decreasing
Atlanta	O_3 (1-hour)	1.4	2	1
	O_3 (8-hour)	-2.5	1	3
	TNMOC	-11.4	0	1
	NO_x	2.1	2	0
Chicago	O_3 (1-hour)	-1.2	1	7
	O_3 (8-hour)	-1.3	2	9
	TNMOC	-7.8	0	2
	NO_x	0.3	1	1
TNMOC = Total nonmethane organic compound				

Methodology

All data were obtained from EPA's AirData Web site (for 1991–2000 data) and AQS database (for 2001 data). Trends are based on data from sites meeting certain data completeness criteria for the 1991–2001 period. Data completeness requirements are the same as those used in previous National Air Quality and Emissions Trends reports.[10] Annual summary statistics for a year of 1-hour or 8-hour ozone data were considered valid if data were available for at least 75 percent of all possible observations. Annual averages computed from round-the-clock 1-hour samples of TNMOC or NO_x were considered valid if data were available for 50 percent or more of all possible observations. For monitors with less frequent TNMOC sampling schedules (1 day in 6, etc.), the annual mean was considered valid if at least 75 percent of scheduled samples were available. Sites selected for trends analysis must have valid annual summary statistics available for 8 or more years for 1991–2001 trends; 5 or more years for 1995–2001 trends. Missing annual summary statistics were filled in via linear interpolation from surrounding years. If a missing value happened to fall at the beginning or end year of the period being investigated, the value was set equal to the nearest available valid year of data.

Composite trends were calculated for each pollutant in both nonattainment areas by averaging the annual summary statistic over all sites in a region. Theil's nonparametric trend-slope estimates and two-sided significance test results for the slope were used to assess statistical significance consistent with the methodology used in previous National Air Quality and Emissions Trends reports. Note that these results are not adjusted for multiple comparisons. Additional methodological details are reported by Coulter-Burke and Stoeckenius.[12]

Rural Area Air Quality Trends

Figure 2-33 presents the trend in 8-hour O_3 concentrations for 34 rural sites from the Clean Air Status and Trends Network (CASTNet) for the most recent 10-year period, 1990–2001.[13] The 8-hour O_3 concentrations at these eastern sites, which were the highest during the hot and dry summers of 1991 and 1998, have decreased 8 percent over the last 10 years. This trend in 8-hour O_3 levels at 34 selected sites is mirrored at other rural sites nationwide. Across the nation, rural 8-hour O_3

levels improved 9 percent from 1981 to 2000, but improved by only 2 percent over the last 10 years.[14]

Figure 2-34 further examines patterns in rural O_3 levels by presenting the 10-year trends in the 8-hour O_3 concentrations at 11 selected National Park Service (NPS) sites.[15] These sites are located in Class I areas, a special subset of rural environments (all National Parks and wilderness areas exceeding 5,000 acres) accorded a higher degree of protection under the Clean Air Act provisions for the prevention of significant deterioration. There are more than 33 NPS sites nationally; however, this analysis focuses on the specific sites with sufficient data to evaluate 10-year trends. Over the last 10 years, 8-hour O_3 concentrations in 33 of our National Parks increased nearly 4 percent. Four monitoring sites in 11 of these parks experienced statistically significant upward trends in 8-hour O_3 levels–Great Smoky Mountains (TN), Mammoth Cave (KY), Yellowstone (WY), and Craters of the Moon (ID). For the remaining 22 parks, 8-hour O_3 levels at 18 increased only slightly between 1992 and 2001, five showed decreasing levels, and three were unchanged.

Figure 2-33. Trends in fourth highest daily 8-hour O_3 concentrations for 34 rural sites from CASTNet, 1990–2001.
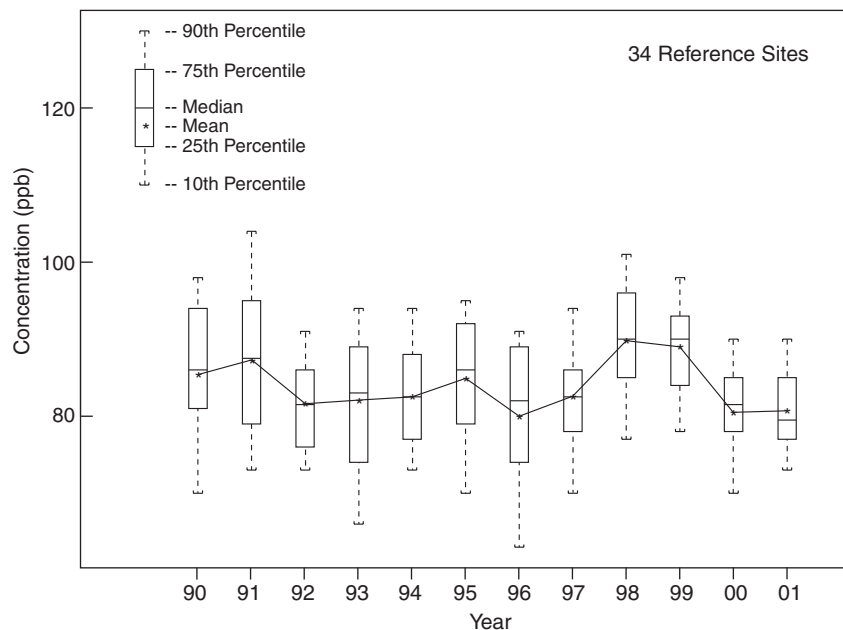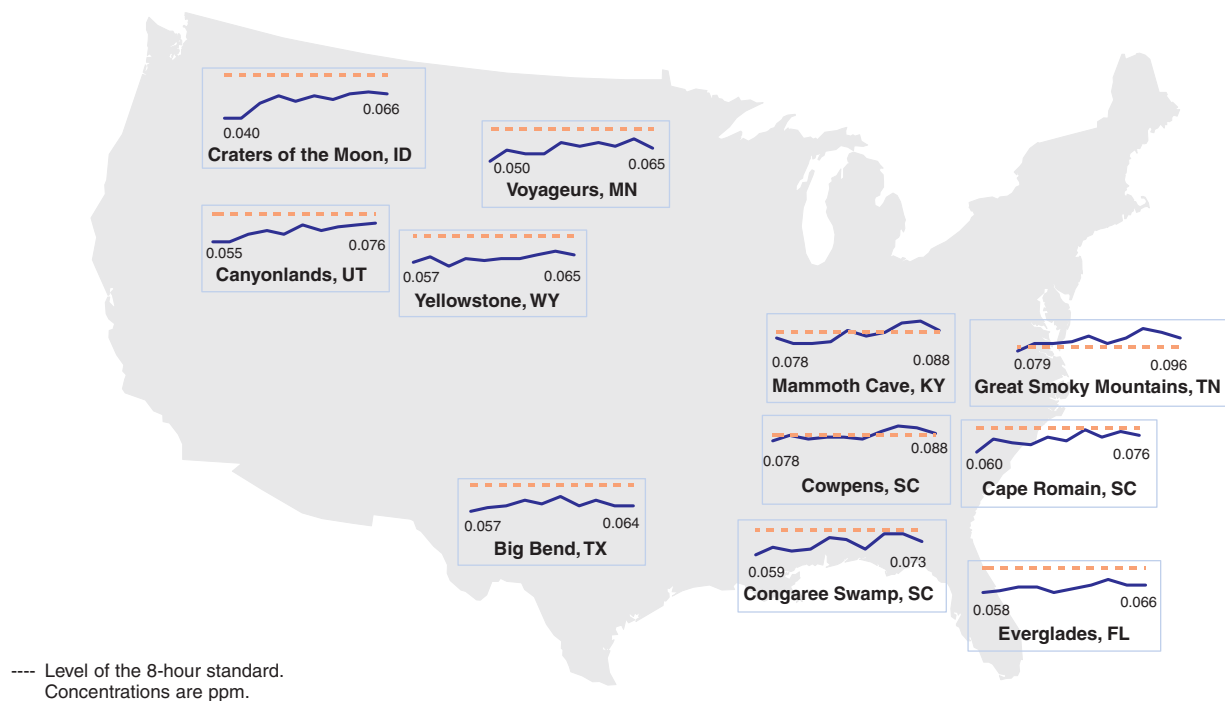

Figure 2-34. Trend in annual fourth highest daily maximum 8-hour O_3 concentrations in National Parks, 1992–2001.

---- Level of the 8-hour standard.
Concentrations are ppm.

National Emissions Trends

Figure 2-35 shows that national total VOC emissions (which contribute to O_3 formation) from anthropogenic (man-made, excluding wildfires and prescribed burnings) sources decreased 40 percent between 1983 and 2002, and 25 percent over the past 10 years. National total NO_x emissions (the other major precursor to O_3 formation) decreased approximately 15 percent and 12 percent, respectively, over the same two periods.

Nationally, the two major sources of VOC emissions are industrial processes (47 percent) and transportation sources (45 percent), as shown in Figure 2-36. Solvent use makes up 63 percent of the industrial processes emission category and 29 percent of total VOC emissions. Industrial process VOC emissions have decreased 26 percent since 1993, in part due to the implementation of maximum achievable control technology (MACT) controls that affect specific chemical and solvent industries. The

VOC emissions totals by source category and year are presented in Table A-5 in Appendix A. Recent control measures to reduce transportation sector emissions include regulations to lower fuel volatility and to reduce NO_x and VOC emissions from tailpipes.[10] The effectiveness of these control measures is reflected in a decrease in VOC emissions from highway vehicles. VOC emissions from highway vehicles have declined 39 percent since 1993, whereas highway vehicle NO_x emissions have decreased 10 percent over the same period.

In addition to anthropogenic sources of VOC and NO_x, there are natural or biogenic sources of these compounds as well. Table 2-5 shows the different predominant plant species responsible for VOC emissions in different parts of the country for two major biogenic species of concern, isoprene and monoterpenes. Although it is not possible to control the level of these natural emissions,

Figure 2-35. VOC emissions, 1983–2002.

1983–02: **40% decrease**[a]
1993–02: **25% decrease**

Note: Emission estimation methods and data sources have evolved over time, resulting in some inconsistency in estimates in different years. In the methods used for this report, the significant changes have occurred between 1984 and 1986 and between 1995 and 1996, although not all source types were affected. More explanation is provided in Appendix B.

[a] Emissions trends data are not available for 1983; thus, the 20-year trend was interpolated based on emissions data for 1980 and 1985.
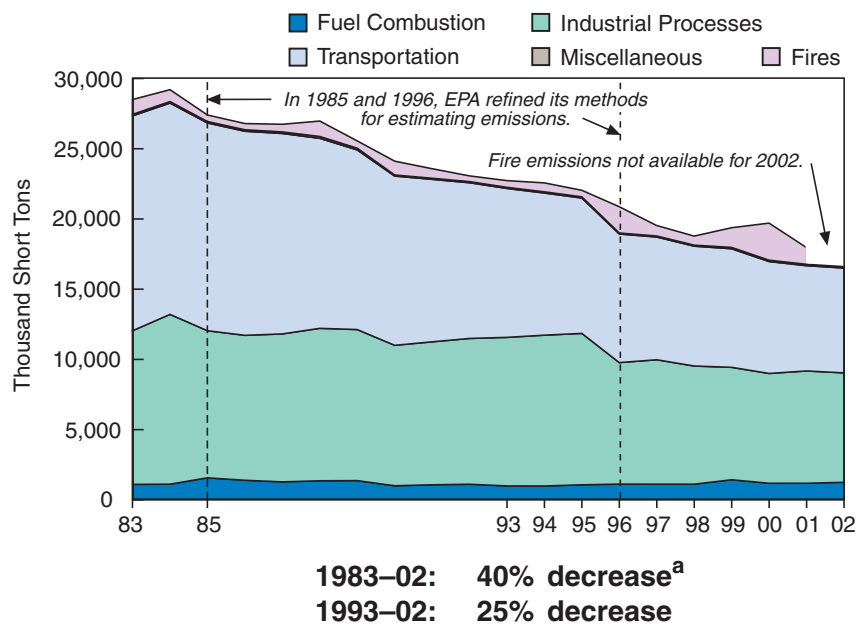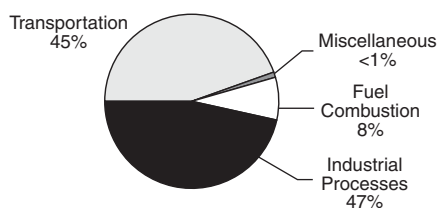
Figure 2-36. Anthropogenic VOC emissions by source category, 2002.[a]

[a] Sums do not equal 100 due to rounding.

Table 2-5. Biogenic Sources of VOC Emissions by Region

Region	VOC	Source
Southwestern United States	Isoprene	Oak (mostly), citrus, eucalyptus
	Monoterpenes	Pine, citrus, eucalyptus
Northeastern United States	Isoprene	Oak (mostly), spruce
	Monoterpenes	Maple, hickory, pine, spruce, fir, cottonwood

their presence is an important factor to consider when developing O_3 control strategies. Biogenic NO_x emissions are associated with lightning and biological processes in soil. On a regional basis, biogenic VOC emissions can be greater than anthropogenic VOC emissions. Biogenic NO_x emissions, however, make up less than 10 percent of total NO_x emissions.[17]

Figure 2-37 shows the geographic distribution of 2001 anthropogenic VOC emissions based on the tonnage per square mile for each county. This map illustrates that the eastern half of the country and the West Coast emit more VOC (on a density basis) than does the western half of the continental United States.

Figure 2-37. Density map of 2001 anthropogenic VOC emissions, by county.

Tons Per Year/Square Mile

- 0.0 - 1.6
- 1.7 - 2.9
- 3.0 - 5.0
- 5.1 - 11
- 12 - 2300
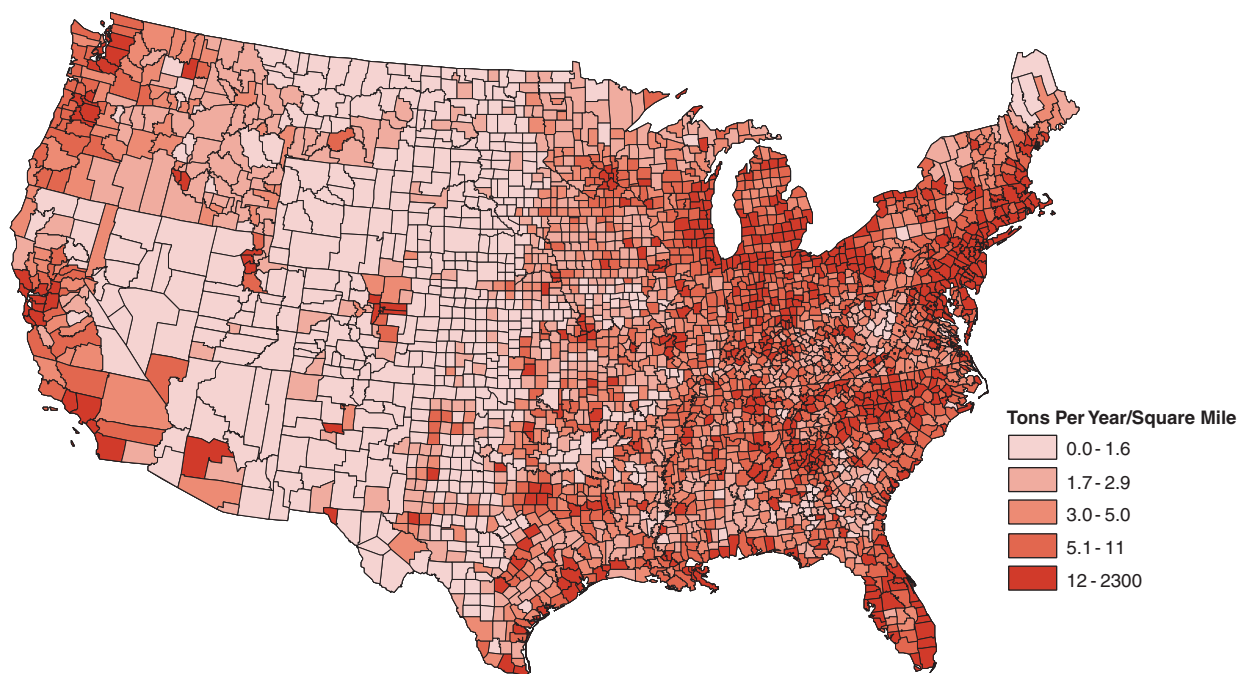
Particulate Matter

PM$_{10}$ Air Quality Concentrations		
1993–02	13%	decrease

PM$_{10}$ Direct Emissions		
1993–02	22%	decrease

PM$_{2.5}$ Air Quality Concentrations		
1999–02	8%	decrease

PM$_{2.5}$ Direct Emissions		
1993–02	17%	decrease

Worth Noting

PM$_{2.5}$

- Annual average PM$_{2.5}$ concentrations decreased 8 percent nationally from 1999 to 2002. The Southeast was responsible for most of that reduction, where the monitored levels of PM$_{2.5}$ decreased 18 percent from 1999 to 2002. Lower 2002 annual average concentrations in the Southeast are due, in part, to decreases in sulfates, which largely result from power plant emissions of SO$_2$.

Nature and Sources

Particulate matter is the general term used for a mixture of solid particles and liquid droplets found in the air. Some particles are large enough to be seen as dust or dirt. Others are so small they can be detected only with an electron microscope. PM$_{2.5}$ describes the "fine" particles that are less than or equal to 2.5 µm in diameter. "Coarse fraction" particles are greater than 2.5 µm, but less than or equal to 10 µm in diameter. PM$_{10}$ refers to all particles less than or equal to 10 µm in diameter. A particle 10 µm in diameter is about one-seventh the diameter of a human hair. PM can be emitted directly or form in the atmosphere. "Primary" particles, such as dust from roads or elemental carbon (soot) from wood combustion, are emitted directly into the atmosphere. "Secondary" particles are formed in the atmosphere from primary gaseous emissions. Examples include sulfates, formed from SO$_2$ emissions from power plants and industrial facilities, and nitrates, formed from NO$_x$ emissions from power plants, automobiles, and other types of combustion sources. The chemical composition of particles depends on location, time of year, and weather. Generally, coarse PM is composed largely of primary particles and fine PM contains many more secondary particles.

Fine and coarse particles typically exhibit different behavior in the atmosphere. Coarse particles can settle rapidly from the atmosphere within hours, and their spatial impact is typically limited because they tend to fall out of the air in the downwind area near their emission point. Larger coarse particles are not readily transported across urban or broader areas because they are generally too large to follow air streams and they tend to be removed easily by impaction on surfaces. Smaller-sized coarse particles can have longer lives and longer travel distances, especially in extreme circumstances, such as dust storms.

Global meteorological conditions play a role in transporting dust periodically from Africa and Asia to North America. A special study, summarized in Chapter 6 and provided in full in the Special Studies section of this report, examines how a particularly large event in Asia in April 2001 affected PM concentrations in the United States.

Health and Environmental Effects

Scientific studies show a link between inhalable PM (alone, or combined with other pollutants in the air), which includes both fine and coarse particles, and a series of significant health effects. Both coarse and fine particles can accumulate in the respiratory system and are associated with numerous adverse health effects. Exposure to coarse particles is primarily associated with the aggravation of respiratory conditions such as asthma. Exposure to fine particles is most closely associated with decreased lung function, increased hospital admissions and emergency room visits, increased respiratory symptoms and disease, and premature death. Sensitive groups that appear to be at greatest risk to such PM effects include the elderly, individuals with cardiopulmonary disease such as asthma or congestive heart disease, and children.

Particulate matter also can cause adverse impacts to the environment. Fine particles are the major cause of reduced visibility in parts of the United States, including many of our National Parks. Other environmental impacts occur when particles deposit onto soils, plants, water, or materials. For example, particles containing nitrogen and sulfur that deposit onto land or waterbodies may change the nutrient balance and acidity of those environments so that species composition and buffering capacity change. Particles that are deposited directly onto the leaves of plants can, depending on their chemical composition, corrode leaf surfaces or interfere with plant metabolism. Finally, PM causes soiling and erosion damage to materials, including culturally important objects such as carved monuments and statues.

Primary and Secondary PM Standards

The NAAQS for PM_{10} were established in 1987. The primary (health-based) and secondary (public welfare-based) standards for PM_{10} include both short- and long-term NAAQS. The short-term (24-hour) standard of 150 $\mu g/m^3$ is not to be exceeded more than once per year, on average, over 3 years. The long-term standard specifies an expected annual arithmetic mean not to exceed 50 $\mu g/m^3$ averaged over 3 years.

The NAAQS for $PM_{2.5}$ were established in 1997. The primary and secondary standards for $PM_{2.5}$ are set at 15 $\mu g/m^3$ and 65 $\mu g/m^3$, respectively, for the annual and 24-hour NAAQS.[18] Compliance with the annual standard is determined by the average of three consecutive annual average values (e.g., for 1999, 2000, and 2001). Compliance with the 24-hour standard is determined by the 3-year average of annual 98th percentile concentrations.

National 10-Year PM_{10} Air Quality Trends

Because 1988 represents the first complete year of PM_{10} data for most monitored locations, a 20-year trend is not available. However, as Figure 2-38 illustrates, the most recent 10-year period (1993 to 2002) shows that the national average of annual mean PM_{10} concentrations at 804 monitoring sites decreased 13 percent. The downward trend is apparent through 1998. However, between 1998 and 1999, the national average increased 1 percent. This slight increase was largely influenced by higher concentrations in the West, particularly in California. PM_{10} concentrations in California were higher than normal from September to December 1999, a period that

coincided with major wildfires and particularly dry conditions.

When the sites are grouped as rural, suburban, and urban, as in Figure 2-39, the individual trends are similar to the national trend. The highest values are generally found at the urban sites, followed closely by

the values at suburban sites. The annual mean is much lower at the rural sites, which are generally located away from local sources of PM_{10}.

Several factors have played a role in reducing PM_{10} concentrations. Where appropriate, states required emissions from industrial sources

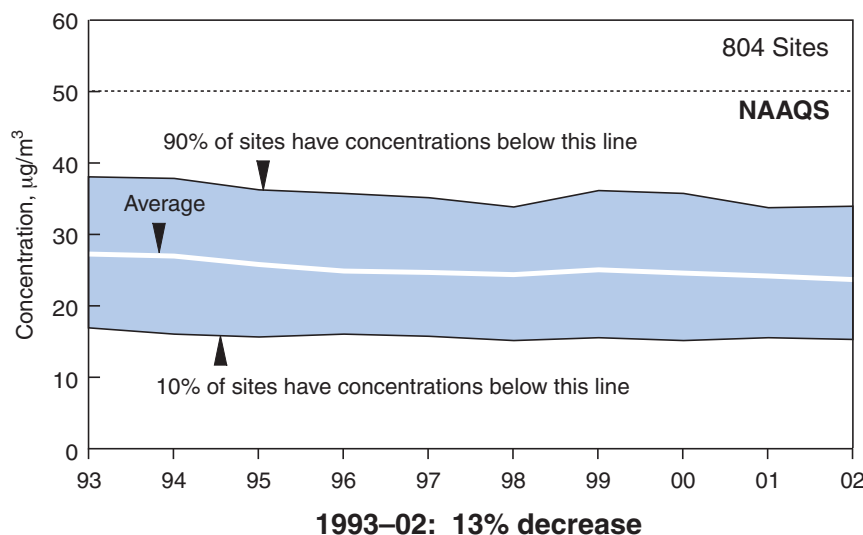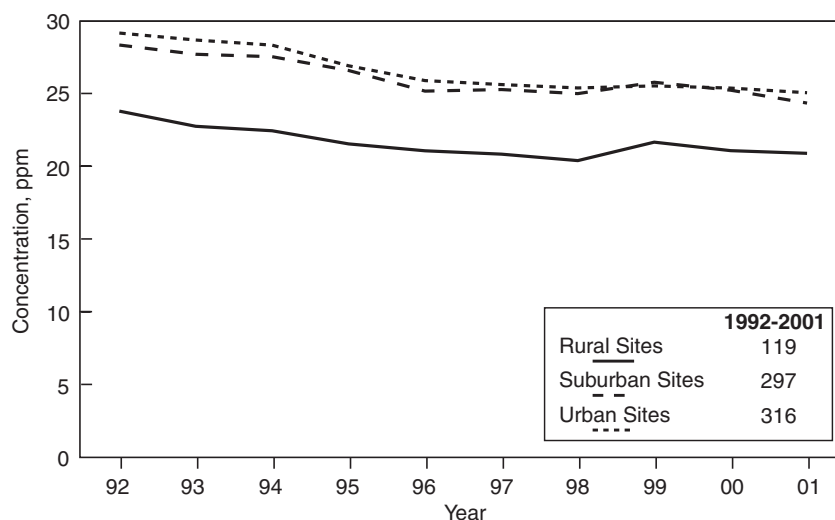**Figure 2-38.** PM_{10} air quality, 1993–2002, based on seasonally weighted annual average.

1993–02: 13% decrease

Figure 2-39. PM_{10} annual mean concentration trends by location, 1992–2001.

	1992-2001
Rural Sites	119
Suburban Sites	297
Urban Sites	316

and construction activities to be reduced to meet the PM_{10} standards. Measures were also adopted to reduce street dust emissions, including the winter-time use of clean anti-skid materials such as washed sand, better control of the amount of material used, and removal of the material from the street as soon as the ice and snow melt. Additionally, cleaner burning fuels such as natural gas and fuel oil have replaced wood and coal as fuels for residential heating, industrial furnaces, and electric utility and industrial boilers.
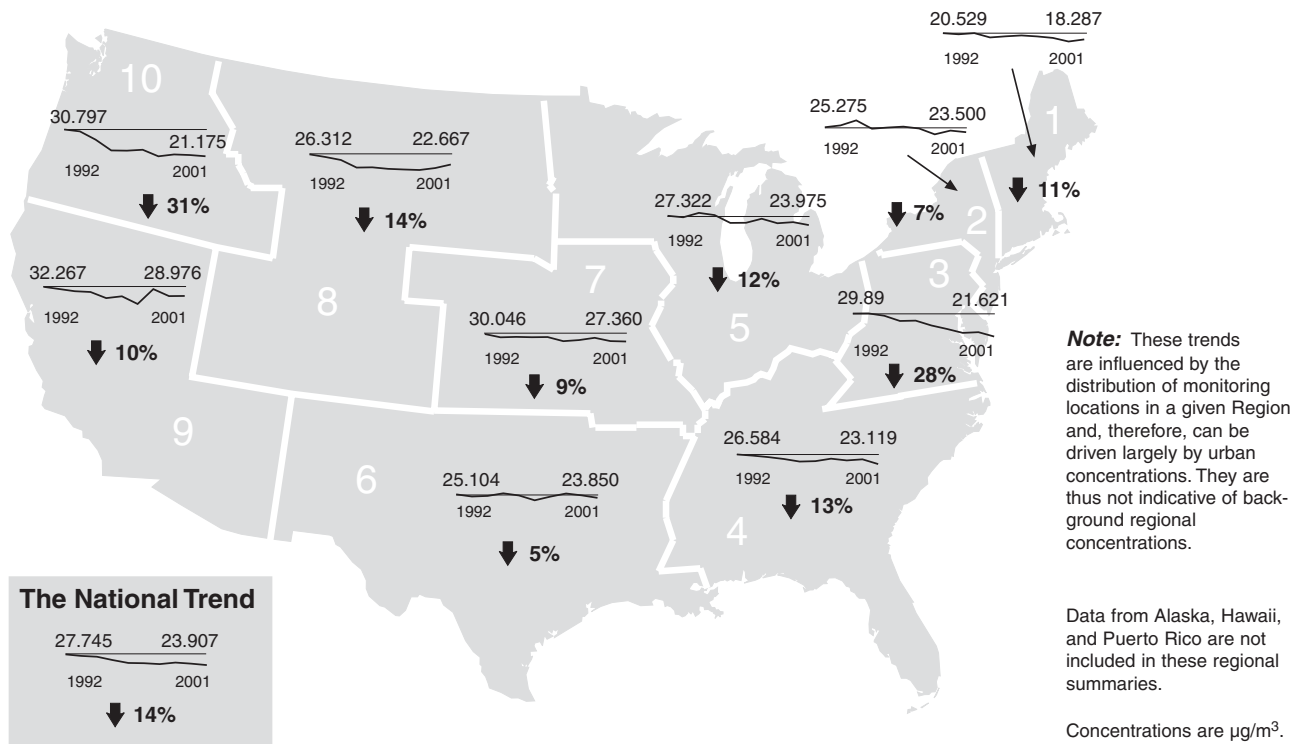
PM_{10} Regional Air Quality Trends

Figure 2-40 is a map of regional trends for the PM_{10} annual mean from 1992 to 2001. All 10 EPA Regions show decreasing trends over the 10-year period, with declines ranging from 5 to 31 percent. The largest 10-year decreases occurred in the Northwest. This is significant because PM_{10} concentrations generally have been higher in the western regions.

In the western States, programs such as those with residential wood stoves and agricultural practices

have helped reduce emissions of PM_{10}.

In the eastern United States, the Clean Air Act's Acid Rain Program has contributed to the decrease in PM_{10} emissions. The program has reduced SO_2 and NO_x emissions, both of which are precursors of particulate matter in the atmosphere (see the SO_2 section in this chapter for more information on the Acid Rain Program).

Figure 2-40. Trend in PM_{10} annual mean concentration by EPA Region, 1992–2001.

Note: These trends are influenced by the distribution of monitoring locations in a given Region and, therefore, can be driven largely by urban concentrations. They are thus not indicative of background regional concentrations.

Data from Alaska, Hawaii, and Puerto Rico are not included in these regional summaries.

Concentrations are µg/m³.

PM_{10} 2001 Air Quality Status

The map in Figure 2-41 displays the highest second maximum 24-hour PM_{10} concentration in each county for 2001. The highest of these was recorded in Inyo County, California, caused by wind-blown dust from a dry lake bed.[19] The bar chart that accompanies the national map shows the number of people living in counties within each concentration range. The colors on the map and bar chart correspond to the colors of the concentration ranges displayed in the map legend. In 2001, approximately 8 million people lived in 13 counties where the highest second maximum 24-hour PM_{10} concentration was above the level of the 24-hour PM_{10} NAAQS. When both the annual and 24-hour PM_{10} standards are considered, there were 11 million people living in 17 counties with PM_{10} concentrations above the NAAQS levels in 2001. See Chapter 4 for information concerning officially designated PM_{10} nonattainment areas.

The Franklin Smelter facility, responsible for historically high recorded PM_{10} concentrations in Philadelphia, shut down in August 1997 and was dismantled in late 1999,[20] resulting in 24-hour concentrations below the level of the standard at the nearby monitoring site.

National PM_{10} Emissions Trends

Direct PM_{10} emissions are generally examined in two separate groups. First, there are the emissions from the more traditionally inventoried sources, which decreased 22 percent nationally between 1993 and 2002 (see Figure 2-42). These sources include fuel combustion, industrial processes, and transportation. Of these, the fuel combustion category saw the largest decrease over the 10-year period (27 percent).

The second group of direct PM_{10} emissions is a combination of miscellaneous and natural sources, including agriculture and forestry, wildfires and managed burning, and fugitive dust from paved and unpaved roads. Although fugitive dust emissions are large and can adversely affect air quality, they do not transport to more distant areas readily as do emissions from other source types. It should be noted that fugitive dust emissions from geogenic wind erosion have been removed from the emissions inventory for all years, because the annual emission estimates based on past methods for this category are not believed to be representative. As Figure 2-43 shows, these miscellaneous and natural sources actually account for a large percentage of the total direct PM_{10}

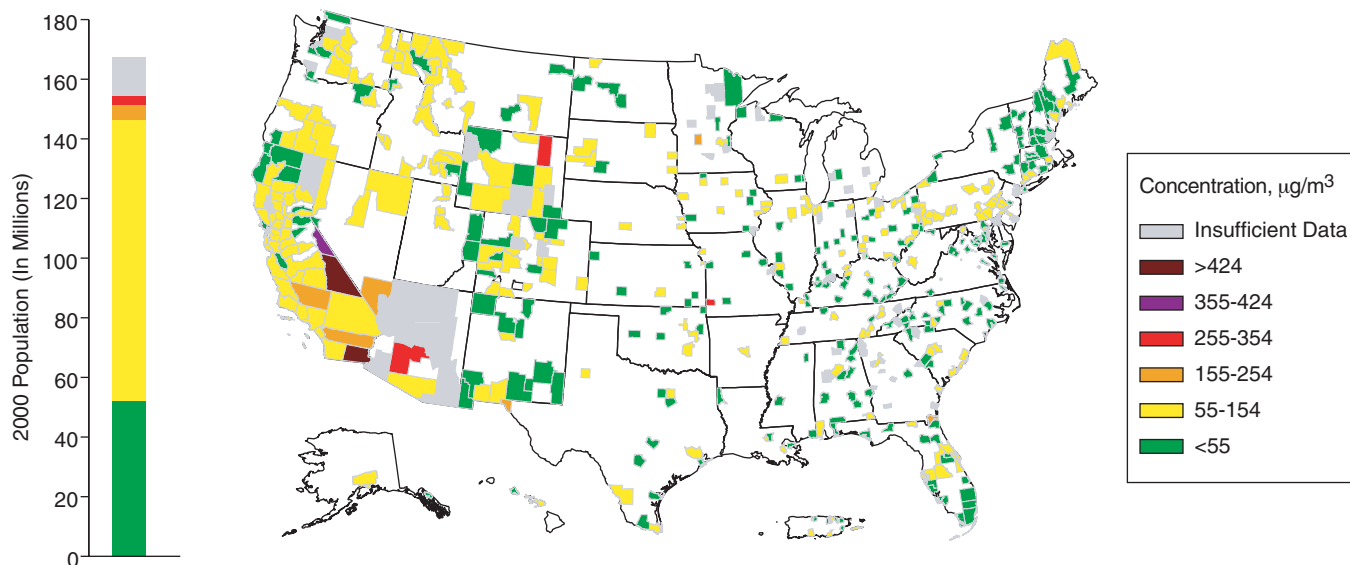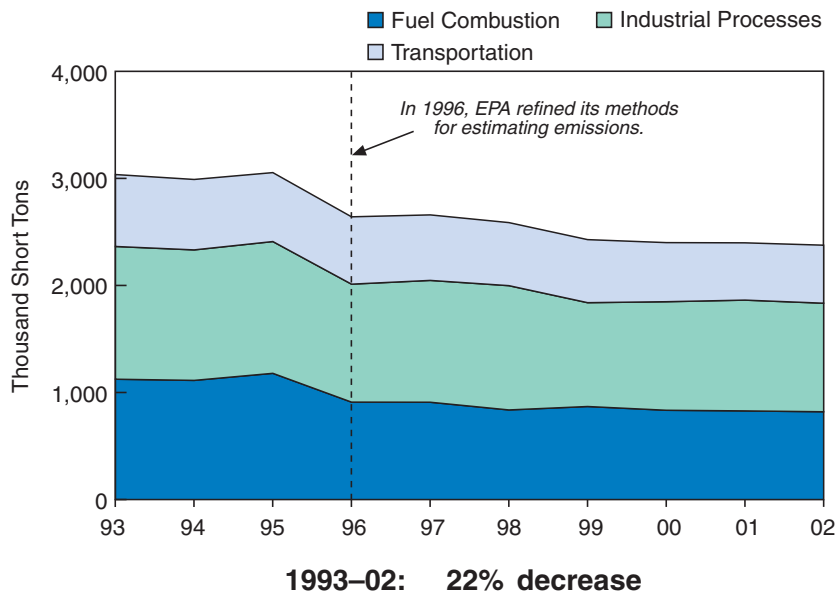**Figure 2-41**. Highest second maximum 24-hour PM_{10} concentration by county, 2001.

Figure 2-42. National direct PM$_{10}$ emissions, 1993–2002 (traditionally inventoried sources only).

1993–02: 22% decrease

Note: Emission estimation methods and data sources have evolved over time, resulting in some inconsistency in estimates in different years. In the methods used for this report, the significant changes have occurred between 1984 and 1986, and between 1995 and 1996, although not all source types were affected. More explanation is provided in Appendix B.

Figure 2-43. National direct PM$_{10}$ emissions by source category, 2002.

emissions nationwide, although they can be difficult to quantify compared to the traditionally inventoried sources. The trend of emissions in the miscellaneous/natural group may be more uncertain from one year to the next or over several years because of this difficulty and because these emissions tend to fluctuate a great deal from year to year.
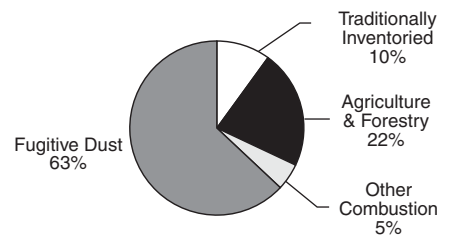
Table A-6 lists PM$_{10}$ emissions estimates for the traditionally inventoried and miscellaneous and natural sources.

Figure 2-44 shows the emission density for PM$_{10}$ in each U.S. county. The PM$_{10}$ emission density closely follows patterns in population density and thus is the highest in the eastern half of the United States, in large metropolitan areas, areas with a high concentration of agriculture (e.g., the San Joaquin Valley in California), and along the Pacific Coast. One exception is that open biomass burning is an important source category that is more prevalent in forested areas and in some agricultural areas. Also, fugitive dust is an important component in arid and agricultural areas.
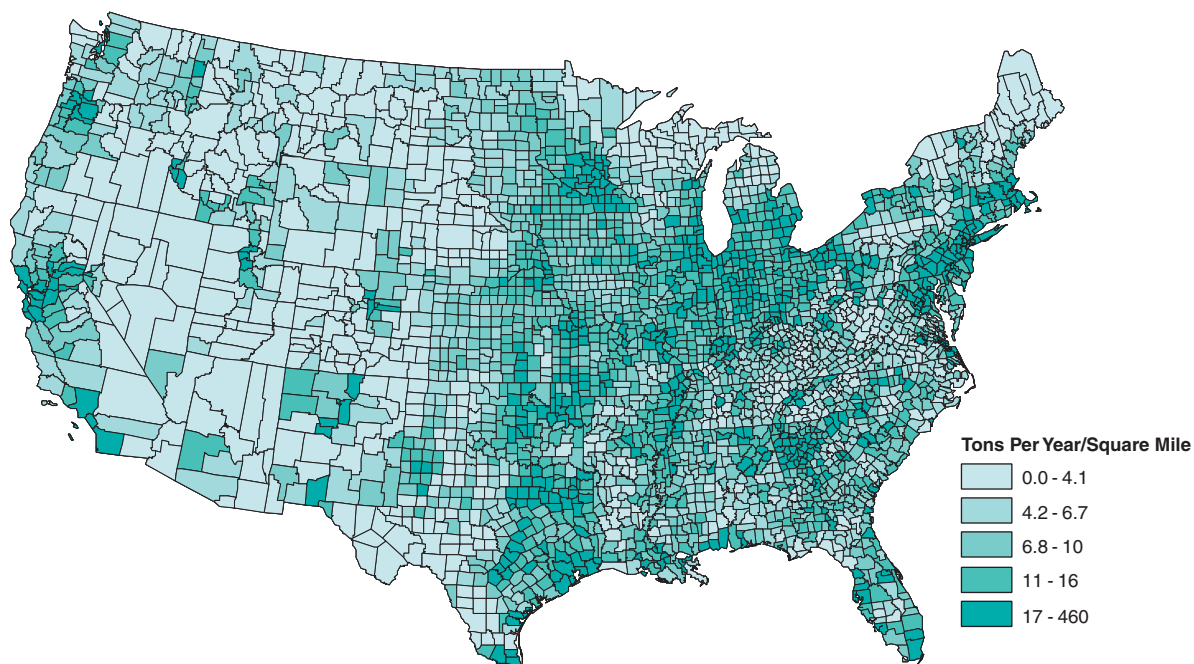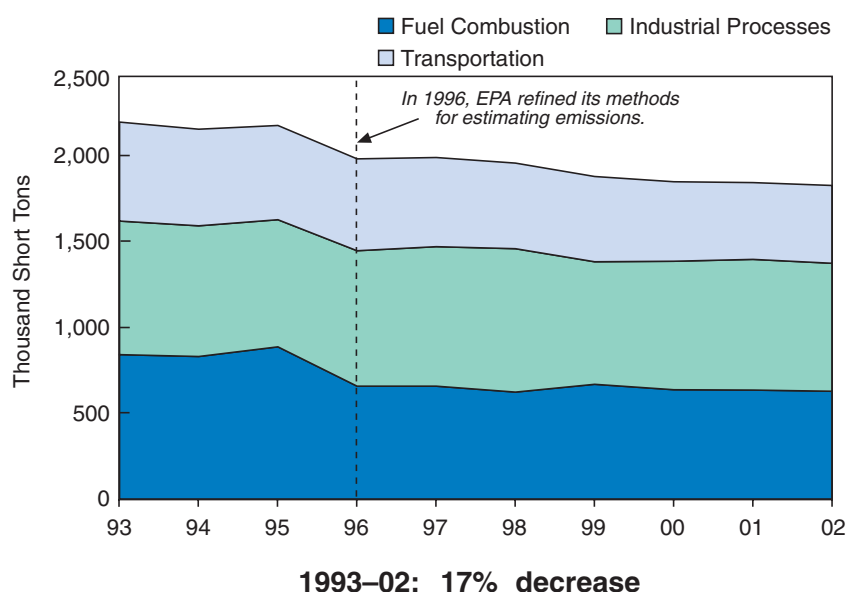
Trends in PM$_{2.5}$ Levels and Direct Emissions

Figure 2-45 shows that direct PM$_{2.5}$ emissions from man-made sources decreased 17 percent nationally between 1993 and 2002. This chart tracks only directly emitted particles and does not account for secondary particles formed when emissions of NO$_x$, SO$_2$, ammonia, and other gases react in the atmosphere. The principal types of secondary particles are sulfates and nitrates, which are formed when SO$_2$ and NO$_x$ react with ammonia.

Figures 2-46 and 2-47 show how sulfates and nitrates, along with other components, contribute to PM$_{2.5}$ concentrations. Figure 2-48 represents the most recent year of data available from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network, which was established in 1987 to track trends in pollutants, such as PM$_{2.5}$, that contribute to visibility impairment. Because the monitoring sites are located in rural areas

Figure 2-44. Direct PM$_{10}$ emissions density by county, 2001.

Tons Per Year/Square Mile

	0.0 - 4.1
	4.2 - 6.7
	6.8 - 10
	11 - 16
	17 - 460

Figure 2-45. National direct PM$_{2.5}$ emissions, 1993–2002 (traditionally inventoried sources only).

■ Fuel Combustion   ■ Industrial Processes
■ Transportation

In 1996, EPA refined its methods for estimating emissions.

Thousand Short Tons

1993–02: 17% decrease

Note: Emission estimation methods and data sources have evolved over time, resulting in some inconsistency in estimates in different years. In the methods used for this report, the significant changes have occurred between 1984 and 1986 and between 1995 and 1996, although not all source types were affected. More explanation is provided in Appendix B.

throughout the country, the network is a good source for assessing regional differences in PM$_{2.5}$. Figure 2-47 represents the most recent year of data from EPA's urban speciation network, which was established in 1999. All of these sites are located in urban areas.

The IMPROVE data show that PM$_{2.5}$ levels in rural areas are highest in the eastern United States and southern California, as shown by the larger circles. Sulfates and associated ammonium dominate the East, with carbon as the next most prevalent component. Sulfate concentrations in the East largely result from SO$_2$ emissions from coal-fired power plants. In California and other areas of the West, carbon and nitrates make up most of the PM$_{2.5}$ measured.

The urban speciation data show that sites in urban areas, as shown in the circles in the map in Figure 2-47, generally have higher annual

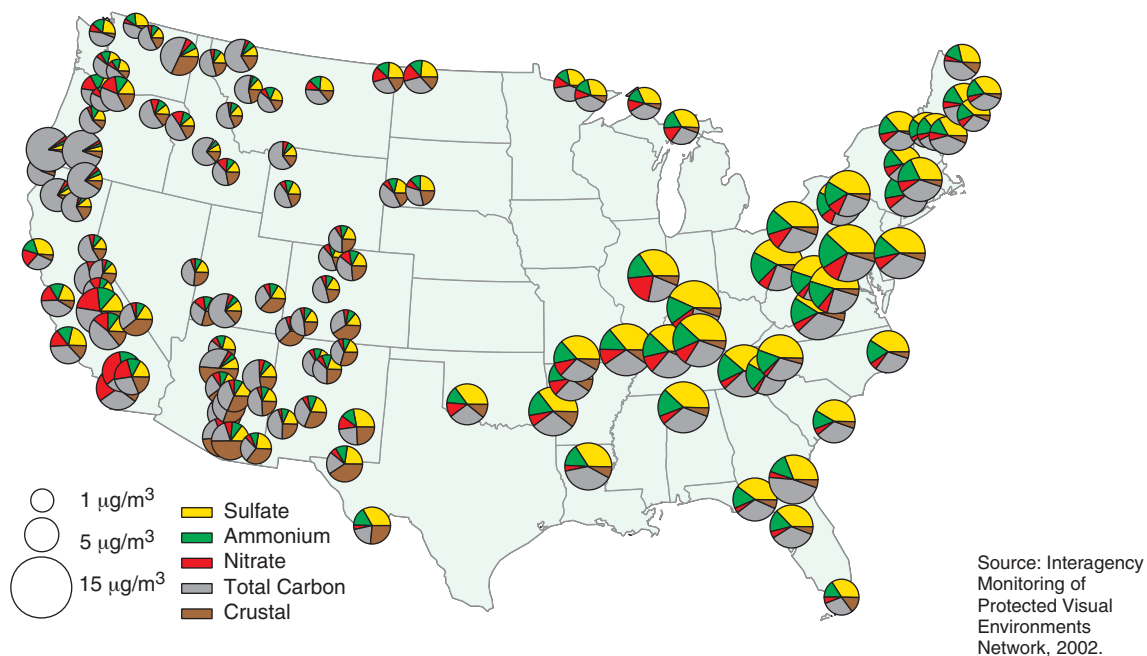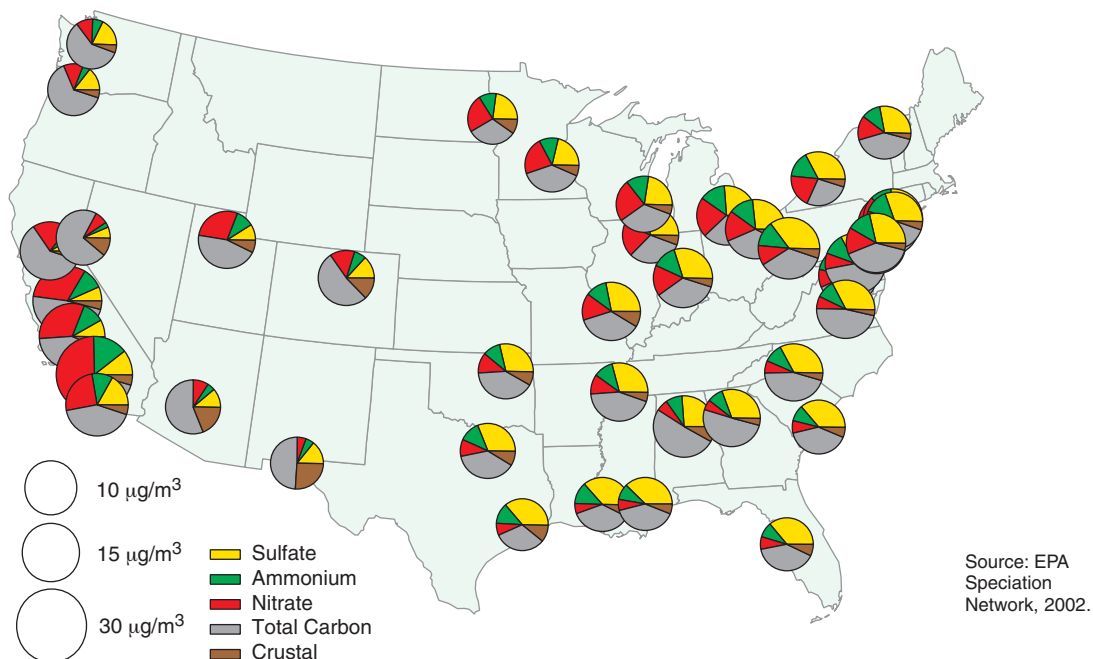**Figure 2-46**. Annual average PM$_{2.5}$ concentrations (µg/m^3) and particle type in rural areas, 2002.

1 µg/m^3
5 µg/m^3
15 µg/m^3

Sulfate
Ammonium
Nitrate
Total Carbon
Crustal

Source: Interagency Monitoring of Protected Visual Environments Network, 2002.

Figure 2-47. Annual average PM$_{2.5}$ concentrations (µg/m^3) and particle type in urban areas, 2002.

10 µg/m^3
15 µg/m^3
30 µg/m^3

Sulfate
Ammonium
Nitrate
Total Carbon
Crustal

Source: EPA Speciation Network, 2002.

Note: Direct comparisons of the information in Figures 2-46 and 2-47 should take into consideration the fact that one is an urban network and the other is a rural network and that there are differences in instruments and measurement methods.

average PM$_{2.5}$ concentrations than nearby rural areas. Urban sites in the East include a large percentage of carbon and sulfates (and ammonium). Urban sites in the Midwest and far West (and especially in California) include a large percentage of carbon and nitrates.

Trends in rural PM$_{2.5}$ concentrations can be examined with data from the IMPROVE network, as shown in Figure 2-48. In the East, where sulfates contribute most to rural PM$_{2.5}$, the annual average PM$_{2.5}$ concentrations decreased 16 percent from 1992 to 2001. This decrease was largely due to a decline in sulfate concentrations, which decreased 17 percent. The other major components remained relatively unchanged over the same period. Average PM$_{2.5}$ concentrations in the West were less than one-half of the average for the eastern sites during this period.

In 1999, EPA and its state, tribal, and local air pollution control partners deployed a monitoring network to begin measuring PM$_{2.5}$ concentrations nationwide. Figure 2-49 shows

annual average PM$_{2.5}$ concentrations by county. This map also indicates that PM$_{2.5}$ concentrations vary regionally. Based on the monitoring data, parts of California and much of the eastern United States have annual average PM$_{2.5}$ concentrations above the level of the annual PM$_{2.5}$ standard, as indicated by the orange and red on the map. With few exceptions, the rest of the country generally

has annual average concentrations below the level of the annual PM$_{2.5}$ health standard.

Now that there are several years of monitoring data available, EPA has begun to examine trends at the national level, as shown in Figure 2-50. Annual average PM$_{2.5}$ concentrations decreased 8 percent nationally from 1999 to 2002. The Southeast was responsible for most

**Figure 2-48**. Annual average PM$_{2.5}$ concentrations in rural areas.
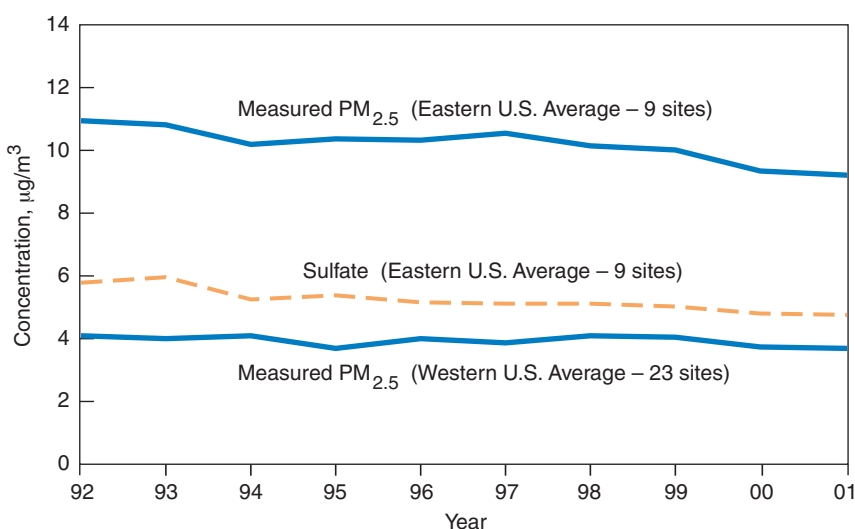
Source: Interagency Monitoring of Protected Visual Environments Network, 1999.

Figure 2-49. Annual average PM$_{2.5}$ concentrations by county, 2001.
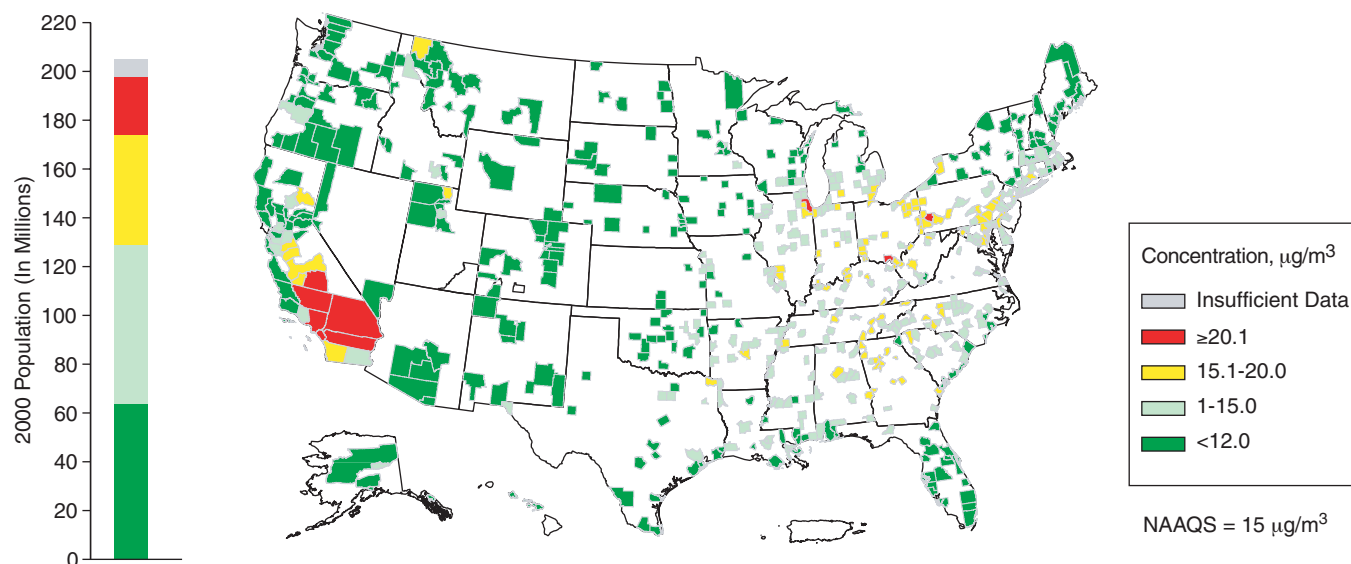
of that reduction, where the monitored levels of $PM_{2.5}$ decreased 18 percent from 1999 to 2002. Lower 2002 annual average concentrations in the Southeast are due, in part, to decreases in sulfates, which largely result from power plant emissions of SO_2.

Figure 2-50. Annual average $PM_{2.5}$ concentrations ($\mu g/m^3$), 2002 (based on seasonally weighted annual average).
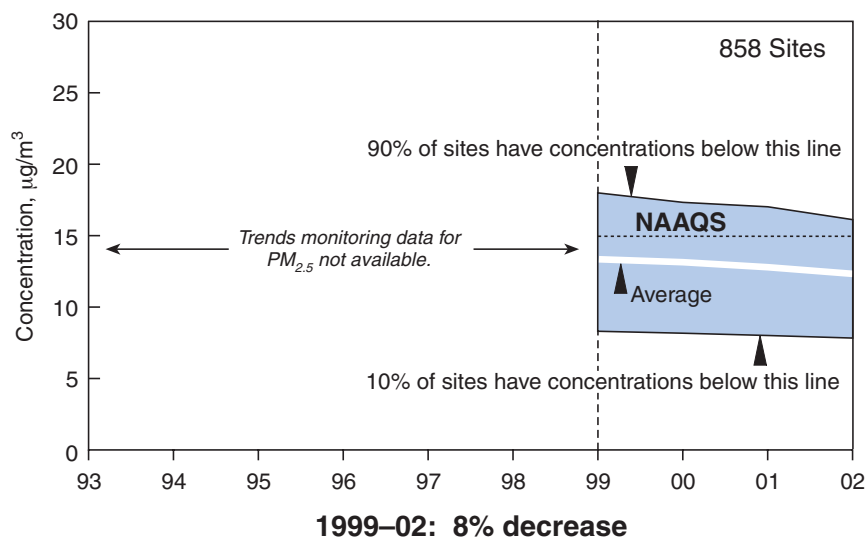
Sulfur Dioxide

Air Quality Concentrations		
1983–02	54%	decrease
1993–02	39%	decrease

Emissions		
1983–02	33%	decrease
1993–02	31%	decrease

Worth Noting

- Steady 20-year improvement has reduced sulfur dioxide (SO_2) ambient concentrations by one-half and emissions by more than one-third.
- Phase II of the Acid Rain Program was implemented in 2000 and has resulted in new reductions.

Nature and Sources

Sulfur dioxide (SO_2) belongs to the family of sulfur oxide (SO_x) gases. These gases are formed when fuel containing sulfur (mainly coal and oil) is burned and during metal smelting and other industrial processes. The highest monitored concentrations of SO_2 have been recorded in the vicinity of large industrial facilities.

Health and Environmental Effects

High concentrations of SO_2 can result in temporary breathing impairment for asthmatic children and adults who are active outdoors. Short-term exposures of asthmatic individuals to elevated SO_2 levels while at moderate exertion may result in reduced lung function that may be accompanied by symptoms such as wheezing, chest tightness, or shortness of breath. Other effects that have been associated with longer-term exposures to high concentrations of SO_2, in conjunction with high levels of PM, include respiratory illness, alterations in the lungs' defenses, and

aggravation of existing cardiovascular disease. The subgroups of the population that may be affected under these conditions include individuals with cardiovascular disease or chronic lung disease, as well as children and the elderly.

Additionally, there are a variety of environmental concerns associated with high concentrations of SO_2. Because SO_2, along with NO_x, is a major precursor to acidic deposition (acid rain), it contributes to the acidification of soils, lakes, and streams and the associated adverse impacts on ecosystems. Sulfur dioxide exposure to vegetation can increase foliar injury, decrease plant growth and yield, and decrease the number and variety of plant species in a given community. Sulfur dioxide also is a major precursor to $PM_{2.5}$ (aerosols), which is of significant concern to human health (as discussed in the particulate matter section of this chapter), as well as a main pollutant that impairs visibility. Finally, SO_2 can accelerate the corrosion of natural and man-made materials (e.g., concrete and limestone) that are used in buildings and monuments, as well

as paper, iron-containing metals, zinc, and other protective coatings.

Primary and Secondary Standards

There are both short- and long-term primary NAAQS for SO_2. The short-term (24-hour) standard of 0.14 ppm (365 µg/m^3) is not to be exceeded more than once per year. The long-term standard specifies an annual arithmetic mean not to exceed 0.030 ppm (80 µg/m^3). The secondary NAAQS (3-hour) of 0.50 ppm (1,300 µg/m^3) is not to be exceeded more than once per year. The standards for SO_2 have undergone periodic review, but the science has not warranted a change since they were established in 1972.

National 10-Year Air Quality Trends

The national composite average of SO_2 annual mean concentrations decreased 39 percent between 1993 and 2002 as shown in Figure 2-51, with the largest single-year reduction (16 percent) occurring between 1994 and 1995.[21] The composite trend has since leveled off, declining only

Figure 2-51. SO_2 air quality, 1983–2002, based on annual arithmetic average.

1983–02: 54% decrease
1993–02: 39% decrease
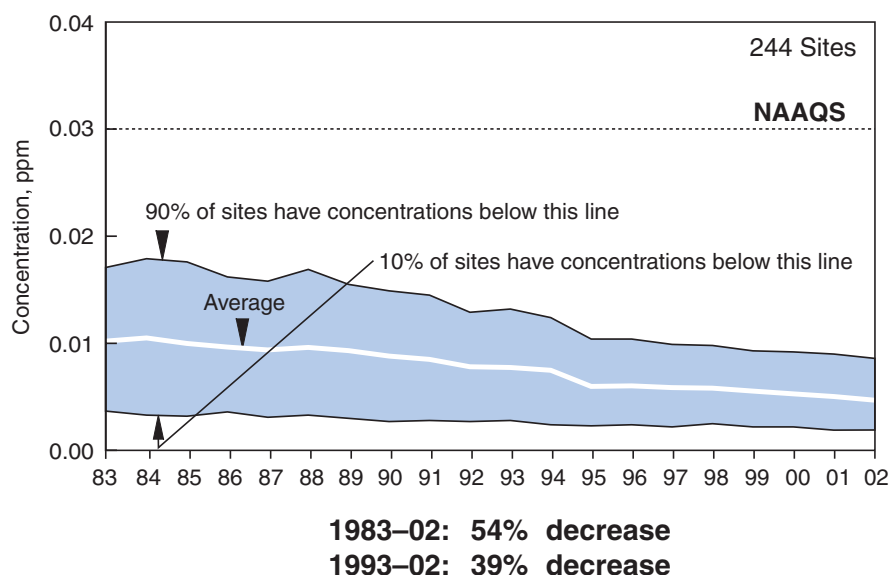
4.5 percent from 2001 to 2002. This same general trend is seen in Figure 2-52, which plots the ambient concentrations grouped by rural, suburban, and urban sites. It shows that the mean concentrations at the urban and suburban sites have been consistently higher than those at the rural sites. However, the 1994 to 1995 reduction in the concentrations at nonrural sites has narrowed the gap between the trends. The greater reduction seen in the nonrural sites reflects the fact that the proportion of nonrural sites is greater in the eastern United States, which is where most of the 1994 to 1995 emissions reductions at electric utilities occurred.[22] The national composite second maximum 24-hour SO_2 annual mean concentrations decreased 35 percent between 1992 and 2001 with the largest single year reduction (25 percent) also occurring between 1994 and 1995.

National Emissions Trends

As shown in Figure 2-53, national SO_2 emissions decreased 31 percent between 1993 and 2002, with an even more impressive 33 percent decrease in the past 20 years (1983 to 2002). The dramatic reduction in 1995 was caused by implementation of the Acid Rain Program; subsequent year-to-year variations are driven in part by the yearly changes in emissions from the electric utility industry, which accounts for most of the fuel combustion category in Figure 2-54. In particular, coal-burning power plants have consistently been the largest contributor to SO_2 emissions, as documented in Table A-9 in Appendix A.

Figure 2-55 shows the emissions density for SO_2 in each U.S. county. SO_2 emissions density is highest in the eastern United States, in large metropolitan areas, and in areas with coal-burning power plants.

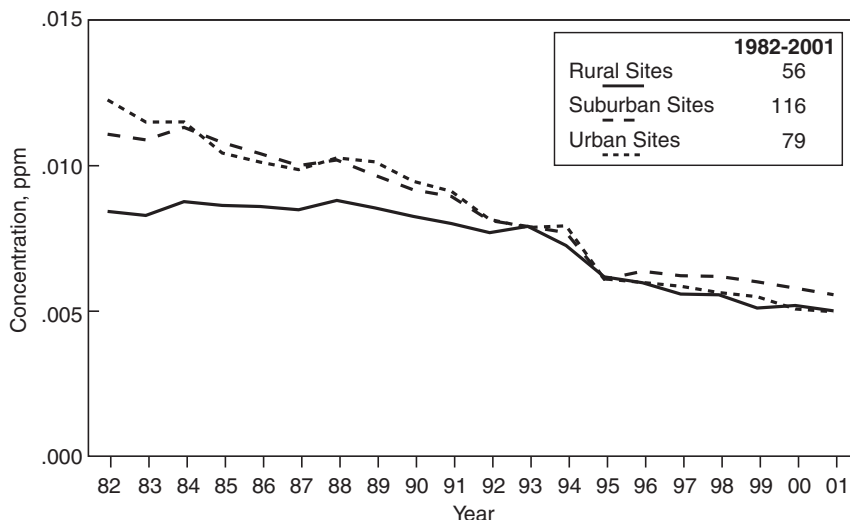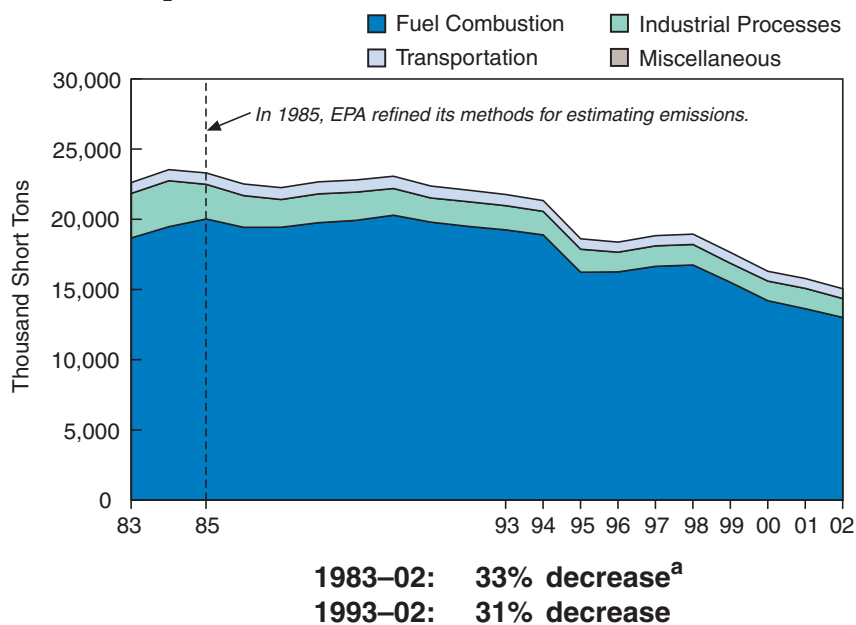**Figure 2-52**. Annual mean SO_2 concentration by trend location, 1982–2001.

Figure 2-53. SO_2 emissions, 1983–2002.

1983–02:   **33% decrease**[a]
1993–02:   **31% decrease**

Note: Emission estimation methods and data sources have evolved over time, resulting in some inconsistency in estimates in different years. In the methods used for this report, the significant changes have occurred between 1984 and 1986 and between 1995 and 1996, although not all source types were affected. More explanation is provided in Appendix B.

[a] Emissions trends data are not available for 1983; thus, the 20-year trend was interpolated based on emissions data for 1980 and 1985.

**Figure 2-54**. SO_2 emissions by source category, 2002.[a]
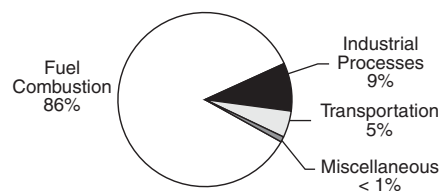
[a] Sums do not equal 100 due to rounding.

Figure 2-55. Direct SO$_2$ emissions density by county, 2001.

Tons Per Year/Square Mile
- 0.0 - 0.12
- 0.13 - 0.29
- 0.30 - 0.80
- 0.81 - 4.3
- 4.4 - 1100
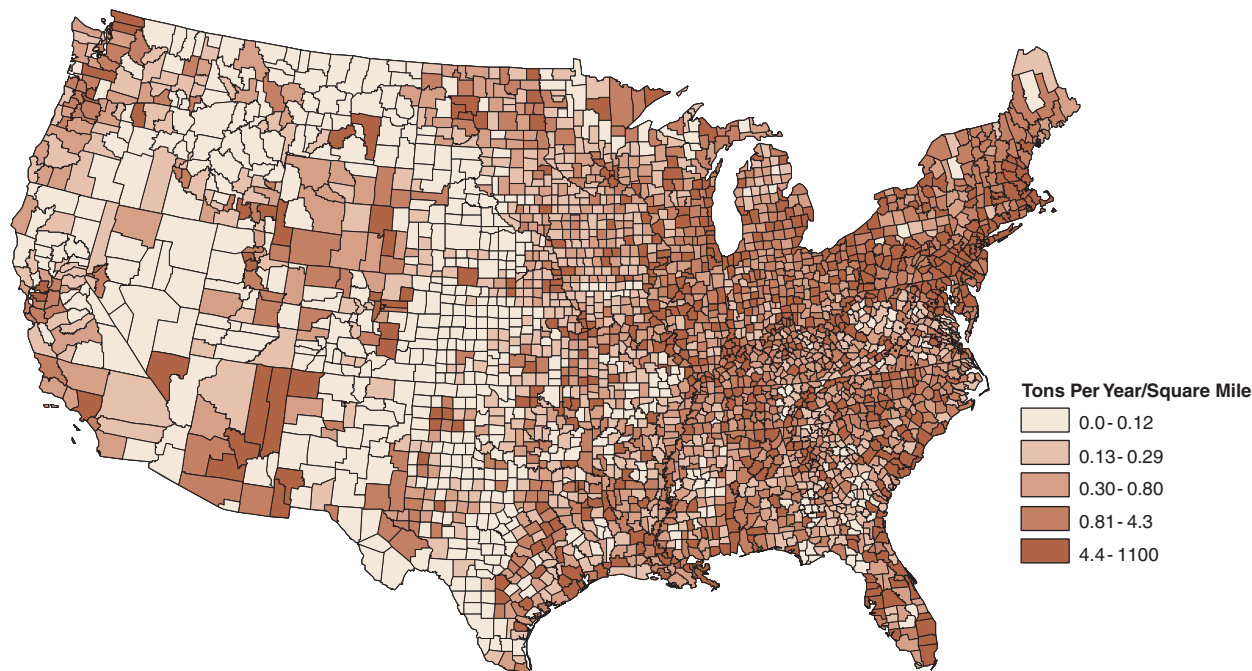
The Acid Rain Program

The substantial national reductions in SO$_2$ emissions and ambient SO$_2$ and sulfate concentrations from 1994 to 1995 were due mainly to Phase I implementation of the Acid Rain Program. Established by EPA under Title IV of the 1990 Amendments, the Acid Rain Program's principal goal is to achieve significant reductions in SO$_2$ and NO$_x$ emissions from electric utilities. Phase I compliance for SO$_2$ began in 1995 and significantly reduced emissions from the participating utilities.[23] Phase II began in 2000 and sets restrictions on Phase I plants as well as smaller coal-, gas-, and oil-fired plants. Approximately 3,000 units are now affected by the Acid Rain Program. Figure 2-56 shows the reduction in SO$_2$ emissions for all sources.

Between 1996 and 1998, total SO$_2$ emissions from electric utilities had increased slightly, compared to their

Figure 2-56. National SO$_2$ emissions trend for all Title IV affected units.
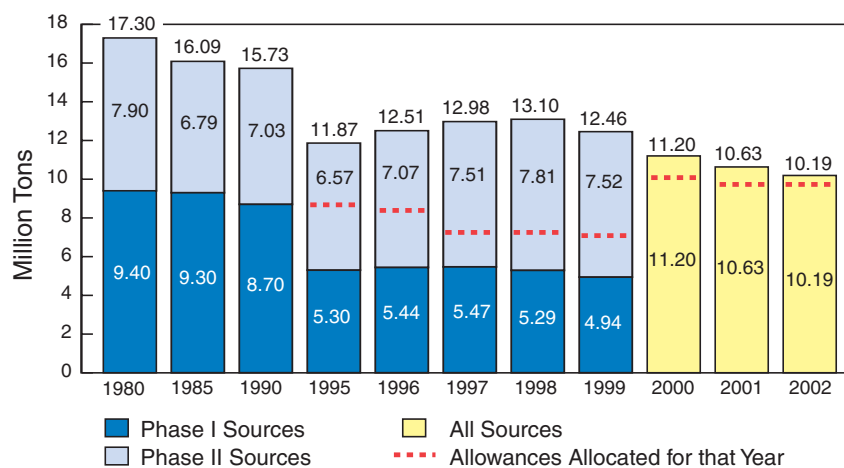
levels in 1995. Since 2000, however, total SO$_2$ emissions have decreased, falling slightly below 1995 levels. Most Phase I plants overcomplied in Phase I (1995 to 2000), banking their

SO$_2$ allowances for use in Phase II, resulting in significant early reductions. However, some Phase I units did increase their emissions during these years. Because Phase I units

account for only 18 percent of the total 1996 to 1998 increase, the majority of the increase is attributed to those units not yet participating in the Acid Rain Program until Phase II. By 2010, the Acid Rain Program will reduce annual SO_2 emissions by half from 1980 levels. The program sets a permanent cap at 8.95 million tons per year on the total amount of SO_2 that may be emitted from power plants nationwide. For more information on the Acid Rain Program, visit **http://www.epa.gov/airmarkets**.

National 20-Year Air Quality Trends

The progress in reducing ambient SO_2 concentrations during the past 20 years is shown in Figure 2-57. The national 2001 composite average SO_2 annual mean concentration is 50 percent lower than it was in 1982. In addition to the previously mentioned effects of the Acid Rain Program, these steady reductions over time were accomplished by installing flue gas control equipment at coal-fired generating plants,

reducing emissions from industrial processing facilities such as smelters and sulfuric acid manufacturing plants, reducing the average sulfur content of fuels burned, and using cleaner fuels in residential and commercial burners.

Regional Air Quality Trends

The map of regional trends in Figure 2-58 shows that ambient SO_2 concentrations are generally higher in the eastern United States. The effects of Phase I of the Acid Rain Program are seen most vividly in the northeast. In particular, concentrations fell 20 to 25 percent between 1994 and 1995 in EPA Regions 1, 2, 3, and 5. These broad regional trends are not surprising because most of the units affected by Phase I of the Acid Rain Program also are located in the East. This figure also shows that ambient concentrations have increased slightly between 1995 and 1997 in Regions 3 and 4 where many of the electric utility units not yet affected by the Acid Rain Program are located.

2001 Air Quality Status

The most recent year of ambient data shows that all counties did meet the primary SO_2 short-term standard, as shown by Figure 2-59.
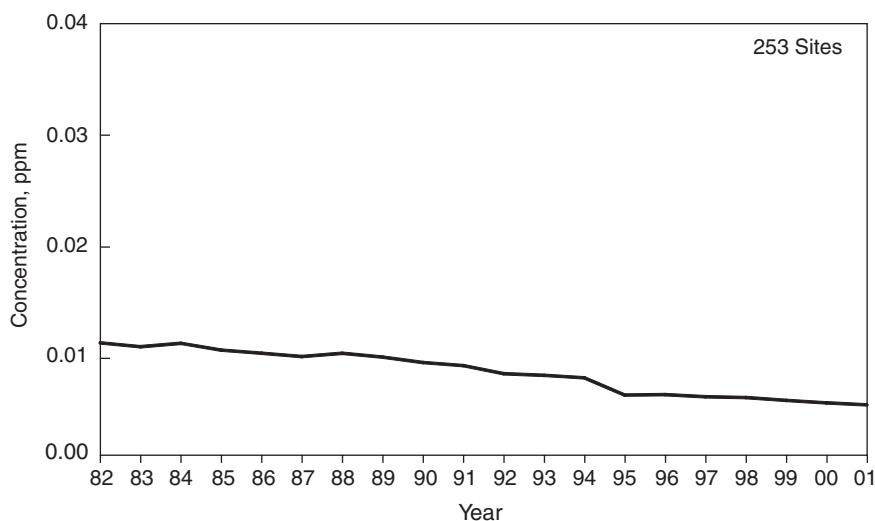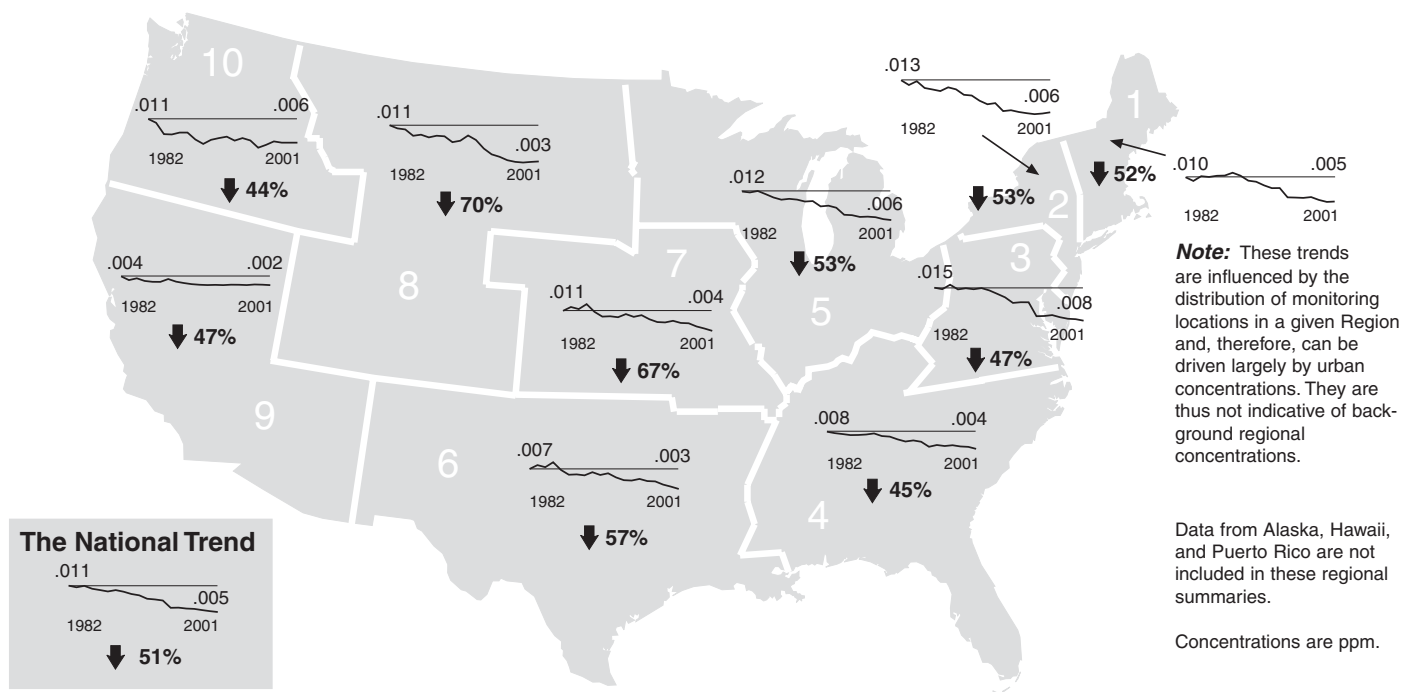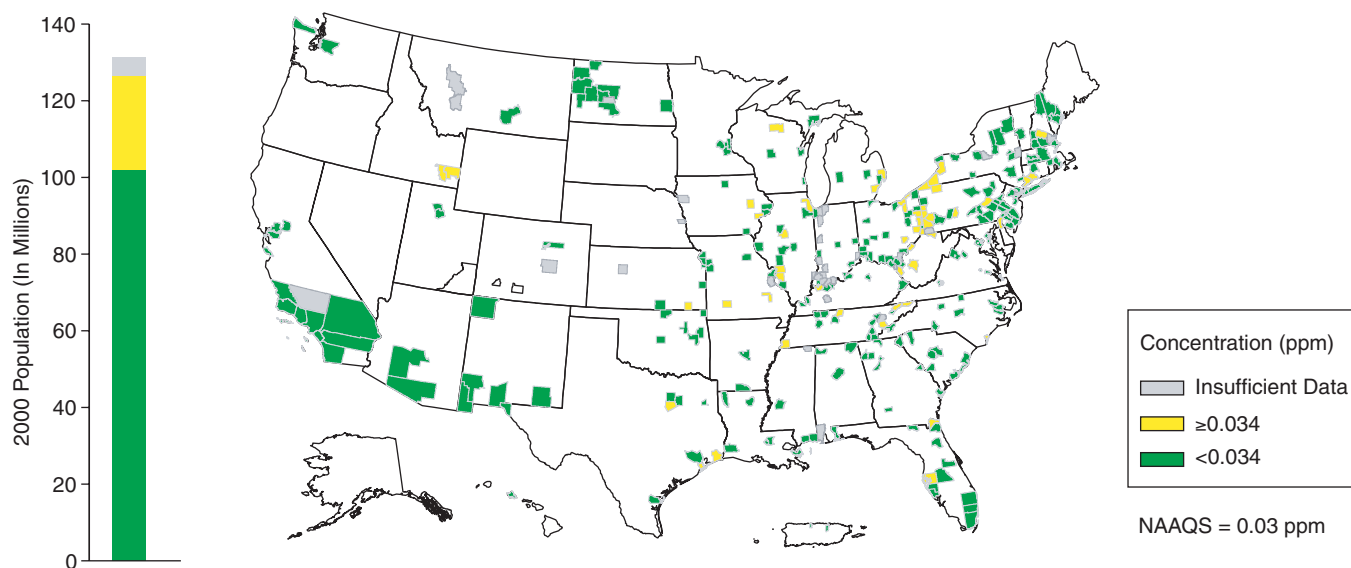
Figure 2-57. Long-term ambient SO_2 trend, 1982-2001.

Figure 2-58. Trend in SO$_2$ annual arithmetic mean concentration by EPA Region, 1982–2001.

Note: These trends are influenced by the distribution of monitoring locations in a given Region and, therefore, can be driven largely by urban concentrations. They are thus not indicative of background regional concentrations.

Data from Alaska, Hawaii, and Puerto Rico are not included in these regional summaries.

Concentrations are ppm.

Figure 2-59. Highest SO$_2$ annual mean concentration by county, 2001.

Concentration (ppm)
- Insufficient Data
- ≥0.034
- <0.034

NAAQS = 0.03 ppm

References

1. Note that due to the annual loss and replacement of ambient monitoring sites (e.g., redevelopment, new leases), too few sites possess a monitoring record sufficient to construct a representative 20-year trend for the nation. Therefore, this report assesses long-term trends by piecing together two separate 10-year trends databases.

2. *Oxygenated Gasoline Implementation Guidelines*, EPA, Office of Mobile Sources, Washington, DC, July 27, 1992.

3. *Guidelines for Oxygenated Gasoline Credit Programs and Guidelines on Establishment of Control Periods Under Section 211(m) of the Clean Air Act as Amended*, 57 FR 47853 (October 20, 1992).

4. Table of winter oxygenated fuels programs by state, EPA, Office of Transportation and Air Quality, Washington, DC, December 8, 1999. **http://www.epa.gov/otaq/regs/fuels/oxy-area.pdf**

5. National Ambient Air Quality Standards for Nitrogen Dioxide: Final Decision, *Federal Register*, 61 FR 196, Washington, DC, October 8, 1996.

6. *Review of the National Ambient Air Quality Standards for Nitrogen Oxides: Assessment of Scientific and Technical Information,* EPA-452/R-95-005, U.S. Environmental Protection Agency, Research Triangle Park, NC, September 1995.

7. Atmospheric concentrations of NO_2 are determined by indirect photomultiplier measurement of the luminescence produced by a critical reaction of NO with ozone. The measurement of NO_2 is based first on the conversion of NO_2 to NO, and then subsequent detection of NO using this well-characterized chemiluminescence technique. This conversion is not specific for NO_2, hence chemiluminescence analyzers are subject to interferences produced by response to other nitrogen-containing compounds (e.g., peroxyacetyl nitrate [PAN]) that can be converted to NO. The chemiluminescence technique has been reported to overestimate NO_2 due to these interferences. This is not an issue for compliance because there are no violations of the NO_2 NAAQS. In addition, the interferences are believed to be relatively small in urban areas. The national and regional air quality trends depicted are based primarily on data from monitoring sites in urban locations and are expected to be reasonable representations of urban NO_2 trends. That is not the case in rural and remote areas, however, where air mass aging could foster greater relative levels of PAN and nitric acid and interfere significantly with the interpretation of NO_2 monitoring data.

8. *1998 Compliance Report*, U.S. Environmental Protection Agency, Acid Rain Program, Washington, DC, August 1999.

9. National Ambient Air Quality Standards for Ozone; Final Rule, *Federal Register*, 62 FR 38856, Washington, DC, July 18, 1997.

10. United States Environmental Protection Agency. Office of Air Quality Planning and Standards. 2000. "National Air Quality and Emissions Trends Reports, 1998." Appendix B.

11. The 1-hour annual ozone design value is defined at an individual monitoring location as the second highest daily maximum 1-hour average concentration; the 8-hour annual design value is defined as the fourth highest daily maximum 8-hour average concentration.

12. Coulter-Burke, S. and T. Stoeckenius, 2002. *Analysis of Ambient Air Quality Trends in the Chicago and Atlanta Ozone Nonattainment Areas.* ENVIRON International Corp., September.

13. CASTNet is considered the nation's primary source for atmospheric data to estimate dry acidic deposition and to provide data on rural ozone levels. Used in conjunction with other national monitoring networks, CASTNet helps to determine the effectiveness of national emission control programs. Established in 1987, CASTNet now comprises 79 monitoring stations across the United States. The longest data records are primarily at eastern sites. The majority of the monitoring stations are operated by EPA's Office of Air and Radiation; however, 27 stations are operated by the National Park Service (NPS) in cooperation with EPA. The CASTNet data complement the larger O_3 data sets gathered by the State and Local Air Monitoring Stations (SLAMS) and National Air Monitoring Stations (NAMS) networks with additional rural coverage.

14. Similarly, although registering declines in 8-hour ozone levels of 16 and 12 percent, respectively, over the last 20 years, urban and suburban site progress slowed between 1991 and 2000 (to 8.5 and 8 percent improvement).

15. This analysis utilizes a nonparametric regression procedure to assess statistical significance, a description of which is provided in Chapter 3: Criteria Pollutants – Metropolitan Area Trends.

16. "Volatility Regulations for Gasoline and Alcohol Blends Sold in Calendar Years 1989 and Beyond," *Federal Register*, 54 FR 11868,

Washington, DC, March 22,1989.

17. *Reformulated Gasoline: A Major Step Toward Cleaner Air*, EPA-420-B-94-004, U.S. Environmental Protection Agency, Office of Air and Radiation, Washington, DC, September 1994.

18. National Ambient Air Quality Standards for Particulate Matter: Final Rule, *Federal Register*, 62 FR 38652, Washington, DC, July 18, 1997. **http://www.epa.gov/ttn/oarpg/t1/fr_notices/pmnaaqs.pdf**.

19. Personal communication with EPA Region 9.

20. Personal communication with EPA Region 3.

21. *Revised Requirements for Designation of Reference and Equivalent Methods for PM2.5 and Ambient Air Quality Surveillance for Particulate Matter: Final Rule*, *Federal Register 62* July 18, 1997.

22. IMPROVE, Cooperative Center for Research in the Atmosphere, Colorado State University, Ft. Collins, CO, May 2000.

23. *1997 Compliance Report: Acid Rain Program*, EPA-430-R-98-012, U.S. Environmental Protection Agency, Office of Air and Radiation, Washington, DC, August 1998.